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THE TRANSITION PERIOD

FOLLOWING THE REPLACEMENT of the McMahon Act by new domestic legislation, the first concern will be to avoid a period of confusion and stagnation such as that which characterized the entrance of the present administration.

The new legislation, which was described in the July issue of NUCLEONICS, decentralizes an important and completely centralized area of our industrial economy. This is a substantial change, and it is inevitable that it will be felt. However, the change from entirely government-controlled to largely privately-initiated development is expected to be extended over a period of years, and, consequently, the immediate dislocations need be no greater than those normally associated with any change of administration.

We shall, therefore, consider the immediate steps which will be associated with the transfer of authority, with a view to assuring continuity of all present operations, while at the same time preparing for a new burst of activity. The problem naturally divides itself into two parts, one having to do with permanent structures of the Commission, such as national laboratories and production plants now in existence, and the other with current projects.

Normal procedures are available for the vigorous continuance of projects now under way. The funds to cover existing construction-contract commitments (largely for fissionable-material production plants) have already been appropriated by Congress. There is no thought that these should be interrupted before completion. As a matter of fact, the AEC took over contracts from the Manhattan District in exactly this way. From a more positive standpoint, we may expect that in the long run fissionable material will be produced from privately owned breeders which are operated for power and production purposes, or simply as converters. Should further fissionable-material production be required before the economics of nuclear power are clear enough to induce private investors to risk their own capital, the Coordinating Board would then assign the Production Corporation the responsibility for producing this additional fissionable material. The Production Corporation could then proceed either to build a government-financed plant by contract, as is now being done, or by some other means.

Gradually it will be possible to turn more and more operations from private operation and government ownership to private operation and private ownership. There is no doubt that certain other spheres of activity, such as mining and production of special materials and chemicals, can be immediately freed from government monopoly.

We shall next turn to the disposition of the permanent facilities of the Commission. These functions and facilities will be apportioned among the new

AEC, the Production Corporation, and the military. Of course, some properties such as mining properties and chemical plants will not go to any of these three but will revert to the public by sale at public auction through the War Assets Administration.

The Argonne, Oak Ridge, Brookhaven, Berkeley, and Los Alamos laboratories will, naturally, become part of the new AEC. In our view, the Los Alamos Scientific Laboratory will function as the weapons research and development center of the new AEC. The assignment of Los Alamos to the AEC instead of to the military is considered expedient inasmuch as the scientists at this laboratory have repeatedly expressed their unwillingness to work for the military. The Knolls Atomic Power Laboratory also would become a development facility of the new AEC. It should be a key part of the AEC program for development of nuclear reactors. Production and distribution of radioisotopes will remain one of the AEC's essential functions.

When, as is to be expected and hoped for, a National Science Foundation is established, most of the long-range research (cosmotrons, meson-theory, cosmic-rays, and other fundamental physical, chemical, and biological research) will fall into its province.

The major fissionable-material production facilities and the plants for processing raw materials will fall under the jurisdiction of the Production Corporation. This includes K-25, Y-12, and the Hanford production units. The Mound Laboratory in Miamisburg, the Ames Laboratory, and the quality-control laboratory in New Brunswick, will also be assigned to the Production Corporation. We may assume that those parts of the AEC administrative apparatus which are concerned with production of fissionable material or source materials will also go to the Production Corporation.

Those parts of the present central administration which will be released to military initiative are also easily named. The NEPA Project will logically become the responsibility of the Air Force, and the Nuclear Energy for the Propulsion of Ships Project will become the responsibility of the Navy. The stockpile of assembled bombs, which will be a fraction of the total fissionable-material stockpile determined by the Coordinating Board, will be the responsibility of the Army Engineers, as will also be the testing grounds at Eniwetok.

We are next faced with the thorny problem of the government-owned-and-operated company towns such as those at Hanford and Oak Ridge. These towns operate at a loss and are supported by government subsidy. This subsidy, in the form of below-cost housing and municipal services, represents a hidden part of the salary of the inhabitants. In normal towns, employees eventually come to own considerable equities in their homes, which act as a reserve of capital and enable freedom of movement. From the standpoint of the individual, this hidden part of his salary, which he does not control, seems attractive on a short-term basis. On a long-term basis, however, it deprives him of his self-reliance and bargaining power, and encourages uneconomic habits.

We propose that the government sell its properties to the individual tenants on favorable terms as a first priority and then to others if this is not taken

up. It may be necessary to raise the employees' salaries to make up for this new charge on them. In any event the cost to the government will be less than the deficit which it has to meet to support these towns. The inefficiency of these towns is now notorious because there is no incentive for anyone to feel individually responsible for the cost of community services.

Under the new legal framework, it is clear that many more people will be involved in atomic energy than at present. This sudden expansion of activity will require more efficient dissemination of information and, at the same time, will require that security procedures be adapted to the new circumstances. A Joint Information Board should, therefore, be set up and be responsible for making full information available to all qualified participants.

The emphasis in security will be on accomplishment. There is no disagreement in any informed circles concerning the self-defeating consequences of the present exaggerated secrecy policies. It is only the antagonism between the Commission, on the one hand, and Congress and the Armed Services, on the other, that prevents the Commission from correcting a policy in which, according to its own public statements, it no longer believes.

It is contrary to our philosophy to give a detailed blueprint, or a five-year plan, for the transition period. The principal production installations are run by private contractors, and their operating procedures will be as little affected by new changes in the legal framework as they were by the transfer from the Manhattan District to the Atomic Energy Commission. We have examined the problems in sufficient detail to be convinced that the technical difficulties of the change are not substantial, and are a small price to pay for the immediate benefits and certainly for the long-term gain in productivity.

—Walter M. DeCew

Members of the Editorial Board have asked us to make clear that they do not necessarily endorse the opinions expressed in this month's editorial or in the July editorial entitled "New Legislation to Replace the McMahon Act." This we gladly do. To remove any contrary implication in the term "Editorial Board" we have dropped that designation in favor of "Technical Consultants."

—The Publisher

PRODUCTION OF RADIONUCLIDES*

Radionuclides are produced mainly in the chain-reacting pile and the cyclotron. Reactions for preparing important isotopes are tabulated and discussed. Problems of target preparation and chemical processing of product nuclides are outlined

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RADIONUCLIDES (*1*), produced in chain-reacting piles and cyclotrons, are being used at an ever accelerating rate for isotopic tracing, therapy, and as sources of ionizing radiations in many branches of science, medicine, and engineering. The possibilities of industrial application are being actively explored. This phase of peacetime utilization of nuclear energy is of tremendous current importance and justifies its consideration in a general discussion of nuclear engineering.

General Production Methods

In its capacity to produce large amounts of radioactive materials, the pile has no equal. Radioactive isotopes of all the chemical elements, except helium, can be made with the slow neutrons from a pile by the (n,γ) reaction. Most of these isotopes are available with specific activities high enough to be generally useful. The fission products offer a varied selection of radionuclides, from the middle of the periodic table, in a carrier-free state. These are byproducts of normal pile operation. In addition to the large number of radionuclides produced by these two means, a small group of vitally important nuclides are made, with pile neutrons, by the (n,p) reaction or by a β^- decay chain following an (n,γ) reaction. In this way, large activities of carrier-free preparations of H^3, C^{14}, P^{32}, S^{35},Ca^{45} and I^{131} are made possible (*2*). Since the stable isotopes of these elements all have important biological functions, their use for tracing and therapy is extensive

The cyclotron is the other important machine for the production of radionuclides. Although, in general, it cannot match the production capacity of a pile, it is a more versatile tool. All pile-produced nuclides can be made with a cyclotron, but the main contribution of the cyclotron is a group of radionuclides not made in the pile or not available from the pile with sufficiently high specific activity or isotopic purity for certain types of applications. Some of the nuclides falling into this class are listed in Table 1.

Although other instruments are available for the production of radionuclides, they are of little importance in comparison to the pile and cyclotron. Charged particles from linear accelerators and, to a very limited extent, alpha rays from natural sources can be used to make radionuclides by direct nuclear reactions. These two sources suffer from both comparatively low energy and low intensity. The use of

* This work was supported in part by the Office of Naval Research. The paper will appear as a chapter in "The Science and Engineering of Nuclear Power," Vol. 2 (Addison-Wesley, Cambridge, Mass., in press).

TABLE 1

Some Useful Radionuclides Best Produced by the Cyclotron

Isotope	*Half-life* (3)	*Production Reaction*	*Useful Target*	*Bombardment Location*	*Yield rd/μah* (4)*	*Radiation Measured*	*Alternative Isotopes*
Be^{7}	43 d	Li(*d*,*n*)	Li,LiF $LiBO_2$	ext. probe	low	γ	Be^{10}
C^{11}	20.5 m	B(*d*,*n*)	B_2O_3	ext.	18 (*5*)	β^+	C^{14}
F^{18}	112 m	0(*d*,*n*)	Li_2CO_3 $LiBO_2$	ext. probe		β^+	
Na^{22}	3.0 y	Mg(*d*,α)	Mg MgO	ext. probe	0.065	β^+ or γ	Na^{24}
Si^{31}	170 m	Si(*d*,*p*)	SiO_2	ext.	37	β^-	
Cl^{38}	37 m	Cl(*d*,*p*)	$CaCl_2$	ext.	1,500	β^- or γ	
V^{48}	16 d	Cr(*d*,α)	Cr	probe	low	β^+ or γ	
Mn^{52}	6.5 d	Cr(*d*,2*n*)	Cr	probe	3	β^+ or γ	Mn^{56} Mn^{54}
Mn^{54}	310 d	Fe(*d*,α)	Fe	probe	0.037 (*5*)	γ or X	Mn^{56} Mn^{52}
Fe^{55}	~ 4 y	Mn(*d*,2*n*)	Mn alloy	probe	0.025 (*6*)	X	Fe^{59}
Fe^{59}	44 d	Co(*n*,*p*) (fast neutrons)	Co	Be probe	5×10^{-5}/gm Co (*6*)	β^- or γ	Fe^{55}
Co^{56}	85 d						
Co^{57}	270 d	Fe(*d*,*n*)	Fe	probe	high	β^+ or γ	Co^{60}
Co^{58}	65 d	Fe(*d*,2*n*)					
Zn^{65}	250 d	Cu(*d*,2*n*)	Cu	probe	0.13	γ or X	
As^{74}	16 d	Ge(*d*,2*n*)	Ge alloy	probe	0.074 (*5*)	β^-, β^+, γ	As^{76}
Se^{75}	115 d	As(*d*,2*n*)	As alloy	probe	high	γ or X	
$Kr^{(79,81)}$	34 h	Br(*d*,2*n*)	NaBr	ext.	high	γ	Kr^{85}
Y^{88}	87 d	Sr(*d*,2*n*)	$SrCO_3$	ext.	1.4	γ	
Ag^{106}	8.2 d	Pd(*d*,2*n*)	Pd	probe	moderate	X or γ	Ag^{110}
Xe^{127}	34 d	I(*d*,2*n*)	NaI	ext.	moderate	γ	Xe^{133}
I^{130}	12.6 h	Te(*d*,2*n*)	Te	ext.	32	β^- or γ	I^{131}
Hg^{197}	23 h 64 h	Au(*d*,2*n*)	Au	probe	30 (7)	e^- or X	$Hg^{(203,205)}$

* Yield data are for thick targets and deuteron energies of 14–15 Mev.

Half-life (3)	*Comments*
2.5×10^6 y	
5,100 y	Possibly useful where very high specific activity is necessary for short time
14.8 h	$(n,2n)$ in pile produces very low specific activity Na^{22}
	Short half-life dictates use near source of production
2.6 h 310 d	$\sim 1\%$ of initial activity due to Mn^{54} contamination
2.6 h 6.5 d	
44 d	(n,γ)-Szilard-Chalmers yields high specific activity but mixed isotopes
~ 4 y	
5.3 y	Carrier-free, long-lived isotopic mixture suitable for work where complex decay is unimportant
26.8 h	Activity due in part to 90-d $As^{(72,73)}$ (X-rays) which will not normally be detected
10 y	Fission product Kr^{85} not available yet
225 d	
5.3 d	Fission product Xe^{133} not available yet
8 d	Specific tracer uses where longer-lived isotope not desired
51.5 d	(n,γ) isotope has low specific activity

these two sources for generating neutrons has more general application, but the neutron intensity does not compare favorably with that available from the pile and cyclotron.

High-energy electrons and X-rays from such instruments as the betatron and synchrotron are capable of creating radionuclides but small yields limit their usefulness for production purposes.

Table 2 is a summary of the main types of nuclear reactions. In addition to the ones listed, there is a small but growing number of multiple-particle reactions (*8*). As bombardment energies increase, the probability becomes large for the compound nucleus to lose its energy by emitting more than one particle. Since each proton or neutron lost in this way relieves the excited nucleus of 6–8 Mev of energy, the economy of multiple-particle processes becomes evident. Recent work has extended the range in Z of such reactions as $(d,2n)$ and (d,H^3) (*9*). An ingenious use of the product of the (d,H^3) reaction for initiating secondary reactions such as (H^3,p) has also been reported (*10*).

Production Specifications

For the production of radio nuclides in the carrier-free state or with high specific activity, the important entries in Table 2 are found in the column, ΔZ. Although (n,γ) and (d,p) reactions yield high specific activity products in many instances, the highest specific activity is always obtained when a change of Z occurs. This change in the atomic number means that the product is not isotopic with the target material and chemical separations are possible.

The term "specific activity" is frequently used in discussing the production of radionuclides and tracer studies. The most generally recognized definition is activity per unit weight of the active element. Activity units should express the number of atoms disintegrating per

TABLE 2

Summary of Nuclear Reactions*

Reaction type	*Number of Reactions Giving Radioisotopes*	*Type of Activity of Products*	ΔZ	ΔA
n,γ	128	β^-	0	+1
$n,2n$	82	β^+	0	−1
n,p	66	β^-	−1	0
n,α	40	β^-	−2	−3
n,f	6	β^-	large	large
p,γ	14	β^+	+1	+1
p,n	112	β^+	+1	0
p,α	2	stable	−1	−3
d,n	72	β^+	+1	+1
$d,2n$	28	β^+	+1	0
d,p	121	β^-	0	+1
d,H^3	6	β^+	0	−1
d,α	30	β^+	−1	−2
α,n	65	β^+	+2	+3
α,p	13	stable	+1	+3
γ,n	33	β^+	0	−1
γ,p	4	β^-	−1	−1

* Compiled primarily from Seaborg's tables (*3*).

unit time (second or minute). The most widely used unit is the curie but its proper definition is not always recognized (*11*). Other units are the rutherford (10^6 d/s) (*12*) and most simply, d/s. Weight units should be in terms of the active element (grams or milligrams of P), but are occasionally based on weight of compound (mc/gm NaI) or even ions (mc/mole $PO_4^{=}$).

A carrier-free preparation contains no measurable quantity of stable isotopes of the radioactive element. Since for a given activity the number of atoms present is proportional to the half-life, the weight of active isotope can be readily calculated by the relation:

$$m = 2.4 \times 10^{-24} I A T_{½} \qquad (1)$$

where m = grams of radionuclide, I = activity (d/s), A = mass number, and $T_{½}$ = half-life. For radionuclides with half-lives less than a few years, most preparations will contain weights of a few micrograms or less. For example, a 100-rd sample of P^{32} contains only 9.5×10^{-9} gm of P^{32}.

If the half-life of the radionuclide is of the order of a thousand years or more, 100 rd of a carrier-free preparation may contain anywhere from a few milligrams on up. A 100 rd source of C^{14} weighs about 0.55 mg.

Any definition of high or low specific activity is completely arbitrary but a convenient lower limit for high specific activity material is 10 rd/mg.

The total activity in any preparation of a radionuclide is determined by several factors. These include the intensity and energy of bombarding particles, intrinsic probability of occurrence of a nuclear reaction, amount of target material bombarded, half-life of product nuclide, and the length of bombardment. Thin target yields are given by the equation:

$$I_{(t)} = F\sigma_a N\left(1 - e^{-\frac{0.693t}{T_{½}}}\right) \qquad (2)$$

$I_{(t)}$ = activity, (d/s), after bombardment for time (t); F = flux of bombarding particles; σ_a = reaction probability for activation, a complex function of particle energy; N = number of target atoms; and $T_{½}$ = half-life of product.

Except for slow-neutron irradiations thick-target yields are difficult to calculate because of the complex relationships between target thickness, particle energy, and reaction probability. Graphical methods are generally necessary to relate thin- and thick-target yields for reactions involving high-energy particles. From a purely practical point of view, a direct experimental determination of thick-target yields is often the most simple.

Product purity is an important specification for a radioactive preparation. Specific activity expresses the purity in terms of stable isotopic dilution. Radioisotopic dilution also can be im-

portant, not because of mass but because of activity. The iodine isotope with an 8-day half-life, I^{131}, can be obtained carrier-free in good yields from several reactions. Deuteron bombardment of tellurium and fission of uranium yield this nuclide. However, in both cases, there is a significant contamination from other radioisotopes of iodine such as I^{135}, I^{130}, I^{133}, etc. It is readily obtained isotopically pure from the reactions:

$$Te^{130}(n,\gamma)Te^{131} \xrightarrow[30\ h]{\beta^-} I^{131}$$

Large amounts of I^{131} are produced by this reaction in the Oak Ridge pile.

Radiochemical purity can be attained for any preparation but it must be carefully checked. As this refers to contamination of the desired preparation by non-isotopic radioactive impurities, standard chemical methods can be used for purification. This contamination is derived from two sources. The first is ordinary chemical contamination of the target. Careful choice of target material will minimize this difficulty. A pre-bombardment analysis is an aid in planning the final purification since it serves as a guide as to what elements must be removed.

The products of side reactions in the bombardment constitute the second source of radiochemical contamination. Many of these are present in a carrier-free state and may not be eliminated in normal purification procedures unless stable isotopic carriers are added or scavenging operations used. Scavenging consists of precipitating some material in which the minute quantities of carrier-free contaminant are carried out by adsorption or isomorphous precipitation. Ferric hydroxide, lanthanum fluoride, and manganese dioxide are frequently used non-specific scavengers.

Targets from slow-neutron irradiations are usually more nearly free of radiochemical contamination than cyclotron targets. Less stringent physical specifications for pile targets permit the choice of purer starting materials and side reactions are much less important. The preparation of P^{32} by a probe bombardment in the cyclotron will illustrate the nature of this problem. Radioisotopes of phosphorus, iron, copper, silver, cadmium, zinc (the last three from silver solder used on the target) are present with macroscopic amounts of each element. In addition there are trace quantities of radioisotopes of cobalt, nickel, manganese, indium, and gallium resulting from side reactions. This list does not include possible radionuclides resulting from the bombardment of impurities in the target material.

Chemical contamination of the product can cause serious trouble, particularly with carrier-free preparations. A few milligrams of iron, chromium, or nickel from the stainless steel equipment used in processing P^{32} will cause partial or complete precipitation of the phosphorus when the solution is neutralized. Bacterial and fungal contamination constitute a hazard when radioactive preparations are used in biological and medical work. They also can cause loss of activity by carrying it out of solution by adsorbtive precipitation.

The chemical form in which a radioactive preparation can be obtained depends largely on the ingenuity of the radiochemist. Unfortunately, complex chemical compounds cannot be synthesized and then activated. Only the simplest inorganic compounds can survive a nuclear bombardment without a total loss of the chemical identity of the minute fraction of the molecules containing the atoms activated. Even a simple, slow-neutron capture will impart sufficient recoil energy to the activated atom to break the chemical bonds holding the atom in its molecule.

Unless recombination to form the original molecule can occur, the activated atoms will not appear in molecules of the material bombarded. When a change in atomic number occurs, presynthesis is out of the question but some simple compounds can be made by utilizing the high chemical reactivity of the newly formed atoms.

With (n,γ) reactions under certain special conditions simple organic molecules can be prepared by providing a suitable environment for the activated recoil nucleus. In this way minute amounts of dibromobenzene, methylene iodide, iodobenzene, etc. have been synthesized (*13*).

Production Reactions—Neutrons

Of the several particles used to induce nuclear reactions for the purpose of making radionuclides, neutrons are the most important. Slow neutrons in the pile give rise to (n,γ) and (n,f) reactions and a few important (n,p) and (n,α) reactions. Fast neutrons are useful in making radionuclides by (n,p) and (n,α) reactions in targets with $Z > 20$. Such neutrons are the result of (d,n) reactions in the cyclotron using Be, Li, or B as targets. Beryllium is particularly important because its physical properties make it suitable for high-intensity deuteron bombardment and its reaction probability for (d,n) is high.

Yields of radionuclides from (n,γ) reactions are readily calculated from equation 2. These calculations are valid for thin targets in which the neutron flux is not diminished appreciably in the sample. Thick-source activations can also be calculated. However, neutron activation calculations are generally useful for determining the order of magnitude of activity expected from small samples, and thick-source calculations are unnecessary. In this equation, the reaction probability for slow-neutron activation of the target, σ_a, can be obtained from a compilation of activation cross sections by Seren, Friedlander, and Turkel (*14*) or the Segrè chart (*15*). Table 3 lists some of the more important values of this parameter. The term, F, for neutrons is the slow-neutron flux (nv) which can be determined experimentally for any given setup. To do this, thin foils of some high-cross-section material, such as indium, are irradiated and absolute β-ray measurements made (*14*, *16*). From the known cross section and measured activity of the foils, (nv) can be determined. Typical values for cyclotron neutron irradiations are $(nv) = 10^7$–10^9 $n/cm^2/sec$. From known cross sections (*14*) and available activities of short-lived ma-

TABLE 3

Some Thermal Neutron Cross Sections for Radionuclide Production (*14*)

Radionuclide	*Half-life*	$\sigma_a \times 10^{24}$
Na^{24}	14.8 h	0.63
P^{32}	14.3 d	0.23
S^{35}	87.1 d	0.011
K^{42}	12.4 h	0.067
Ca^{45}	180 d	0.013
Sc^{46}	85 d	22
Cr^{51}	26.5 d	0.5
Fe^{59}	44 d	0.001
Co^{60}	5.3 y	21.7
Cu^{64}	12.8 h	2.0
Zn^{65}	250 d	0.26
As^{76}	26.8 h	4.2
Se^{75}	115 d	0.2
Br^{82}	34 h	1.11
Rb^{86}	19.5 d	0.52
Sr^{89}	55 d	0.0042
Ag^{110}	225 d	1.1
Cd^{115}	43 d	0.04
In^{114}	48 d	2.52
Sb^{122}	2.8 d	3.8
Te^{127}	90 d	0.014
Cs^{134}	1.7 y	25.6
Ta^{182}	117 d	20.6
Au^{198}	2.7 d	96.4
Tl^{206}	3.5 y	2.2
Bi^{210}	5.0 d	0.015

terials (*2*), the value of $(nv) = 10^{11}$–10^{12} $n/cm^2/sec$ can be obtained for the section of the Oak Ridge pile where samples are irradiated.

The values of σ_a depend upon the velocity of the neutrons. In the simplest case, $\sigma_a \propto 1/v$. This is frequently complicated by resonance absorption where the cross section shows a pronounced increase in value for certain values of the neutron velocity. Activation cross sections for fast neutrons are not readily obtained since monoenergetic neutrons are difficult to produce. Some idea of the variations of σ with v can be obtained from the curves of Goldsmith, Ibser, and Feld (*15*). Experimental determination under production conditions is the most reliable method for obtaining fast-neutron yields.

The specific activity of radionuclides produced by the (n,γ) reaction varies widely but in general is low to moderately high.

Through extremely fortunate coincidences, most of the biologically important radionuclides are producible by pile neutrons in (n,p) reactions. These reactions are $N^{14}(n,p)C^{14}$, $S^{32}(n,p)P^{32}$, $Cl^{35}(n,p)S^{35}$, and $Sc^{45}(n,p)Ca^{45}$.

Targets high in nitrogen such as $Ca(NO_3)_2$, NH_4NO_3, KNO_3, and Be_3N_2 have been used for C^{14} production. After irradiation, the carbon is released by dissolving the target. Most targets yield $C^{14}O_2$ which can be recovered by standard techniques involving small volumes of this gas. Some targets give significant quantities of other carbon compounds such as CO, CH_4, HCHO, HCN, CH_3OH, and HCOOH (*17*).

Irradiated metal chlorides generally give $S^{35}O_4^=$ when dissolved in water. Moderate activities (1–3 krd) of S^{35} have been produced by neutron irradiation of CCl_4 with cyclotrons. This production involves approximately a ton of CCl_4 and recovery of the active sulfur as sulfate by hypobromite oxidation of all sulfur compounds to $SO_4^=$ in a two-phase system (*18*). The sulfate can be coprecipitated with $BaCrO_4$ from the large volume of aqueous solution necessary to extract a ton of CCl_4 and is subsequently recovered as pure $BaSO_4$.

When elementary sulfur is irradiated in the pile, large activities of P^{32} can be made. Separation of P^{32} from the bulk sulfur is effected by a two-phase extraction process. At the boiling point of concentrated nitric acid, sulfur is a liquid and the P^{32} concentrates in the acid phase as H_3PO_4 (*19*). A mixture of glacial acetic acid and acetic anhydride can be used instead of concentrated nitric acid (*20*). This latter mixture results in no contamination of the product with H_2SO_4 and a less complex purification procedure is necessary.

The production of Ca^{45} from Sc^{45} is complicated by the simultaneous production of a large amount of Sc^{46}. This radionuclide is long-lived (85 d) and γ-ray emitting. The activity ratio of Ca^{45}/Sc^{46} is of the order of magnitude of 10^{-3}–10^{-4}. The high radiation intensity makes chemical separation hazardous and difficult. The use of specific complexing agents with organic solvent extraction methods has made Ca^{45} available.

For some types of experiments, it is desirable to have Fe^{59} free from Fe^{55}. Both are produced by (n,γ) reactions on iron but $Co^{59}(n,p)Fe^{59}$ yields an isotopically pure product. Unfortunately the yield is low with pile neutrons and the Co^{60} produced simultaneously presents a major radiation hazard. Under normal conditions of pile irradiation, the activity ratio of Fe^{59}/Co^{60} is approximately 7×10^{-5}. Shielding the target with cadium to reduce the (n,γ) reaction in the cobalt raises this ratio to 10^{-3} (*21*). More favorable production of Fe^{59} is by fast neutrons from the cyclotron. Fastening a cadmium-covered piece of cobalt metal to the back of

a beryllium target gives good yields of Fe^{59} (80 μrd per μah of 14 Mev deuterons per gm Co) with an activity ratio of Fe^{59}/Co^{60} of 10^{-1}–10^{-2}.

With the exception of the reactions $Li^6(n,\alpha)H^3$ and $Ca^{40}(n,\alpha)A^{37}$, significant quantities of radionuclides have not been produced by this reaction. It is useful primarily for small quantity production for nuclear chemical studies.

Other fast-neutron reactions such as $(n,2n)$ are not important from a production standpoint.

In addition to its vast importance in the slow-neutron chain reaction and atomic bombs, the (n,f) reaction produces fantastic amounts of radionuclides. These are isotopic with the elements in the middle of the periodic table from Zn to Eu. A complete list of radionuclides produced by this reaction and their relative yields has been published (*22*). Except when produced locally by cyclotron neutrons, only a limited number of fission-product nuclides are available (*2*). The difficulties inherent in rapidly separating a very complex chemical mixture whose radiation intensity is terrifically high prevent a more general distribution of the short-lived fission products.

Two very interesting nuclides from this source have been produced. Elements No. 43 and No. 61 have not been conclusively demonstrated in nature. A long-lived radioisotope of each of these elements has been isolated in milligram amounts and the physical properties of these elements recorded. The names technetium and prometheum have been proposed in recognition of the isolation of macroscopic amounts of Tc^{99} (*23*) and Pm^{147} (*24*). There is considerable evidence that stable isotopes of these elements cannot exist.

Production Reactions—Deuterons

The second most important bombarding particle for the production of radionuclides is the deuteron accelerated to high energies in the cyclotron. Several factors make it favored over protons, He^{++}, and heavier ions. It participates in more different nuclear reactions than the proton and can be accelerated to high energies more readily. Deuterons are more readily produced than He^{++}, thus higher intensity beams are available for bombardment. Their reaction thresholds are generally lower than those for He^{++}.

The most common reaction with deuterons is (d,p). This reaction is completely analogous to the (n,γ) reaction. Since there is no change in Z in this reaction, the products are isotopic with the targets and specific activities run low to moderately high. For short-lived activities (< two weeks), the specific activity readily obtainable by high-energy deuteron bombardment is as high or higher than that obtained by slow-neutron irradiation in the Oak Ridge pile. However, longer-lived nuclides can be built up to higher specific activities with the pile than with the cyclotron because of greater ease in making long bombardments. Although reaction probabilities for deuteron reactions are frequently lower than those for slow neutrons, the deuteron beam has a higher intensity than the neutron flux in the pile.

Reactions such as

$$_{11}Na^{23} + {}_1H^2 = {}_{11}Na^{24} + {}_1H^1 + Q$$

have positive values of Q (reaction energy). These exoergic reactions should occur at low energies but are hindered by the coulomb repulsion resulting from the positive charges on the target nucleus, the deuteron, and the proton. This barrier makes necessary relatively high deuteron energies (1–2 Mev) before appreciable activities are produced.

Thin-target yields for all deuteron reactions can be calculated by equation 2. In this case, $F = d/cm^2/sec = 6.25 \times 10^{12} \times \mu a/cm^2$. The reaction

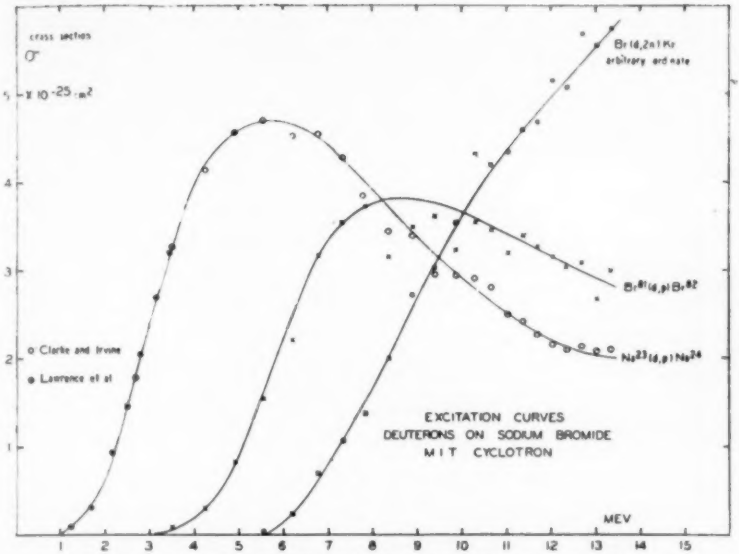

FIG. 1. Excitation curves for deuterons on sodium bromide (*25*)

probability, σ_a, can be determined as a function of deuteron energy. Thick-target yields are obtained by graphical integration of deuteron excitation functions. Details of the experimental methods for determining σ_a and thick-target yields are given by Clarke and Irvine (*25*). Yield values for a few (d,p) reactions are given in Table 1 and in references (*4*, *5*). Measurements of this type are much less complete than for neutron reactions. Figures 1 and 2 give examples of (d,p) and $(d,2n)$ excitation curves (σ_a *vs* E) and of thick-target yields as a function of deuteron energy.

The shape of the (d,p) excitation curve is characteristic for all such curves for this reaction. It starts at moderately low energies, rises to a maximum, and then drops. The decrease in cross section is due to the increasing importance of competing reactions. The thick-target yield curve does not show this decrease. At higher bombarding energies where the (d,p) reaction could become negligible the yield curve would reach a fixed value independent of energy. This is due to increasing range of the deuteron beam in the target with increasing energy.

Although of little importance from the production standpoint, the reaction (d,H^3) can cause trouble in the preparation of pure (d,p) product. Contamination of Co^{60} preparations made with deuterons can amount to as much as 10% of the measured γ-ray activity when thick targets are bombarded with 14-Mev deuterons. Figure 3 shows the shape of the excitation curve for the reaction $Co^{59}(d,H^3)Co^{58}$ (*26*).

For the production of many useful radionuclides in a carrier-free state, the reactions (d,α), (d,n), and $(d,2n)$ are important. The first of these is most productive in the lower half of the

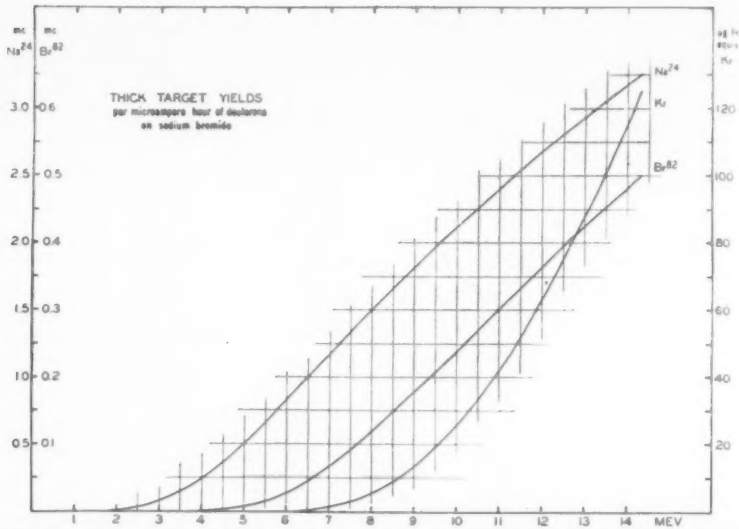

FIG. 2. Thick target yields from deuterons on sodium bromide (25). Ordinates on the left are in millicuries (3.7×10^7 d/s) per microampere hour. Ordinates on the right are in micrograms radium gamma ray equivalent measured through 1.6 mm Pb with a Pt-screen cathode gamma ray counter

periodic table. High values of Z mean a high barrier to the emission of He^{++} from the compound nucleus. Very high energy deuterons are required and the compound nucleus then has sufficient energy so that endoergic reactions such as $(d,2n)$ compete favorably with He^{++} emission.

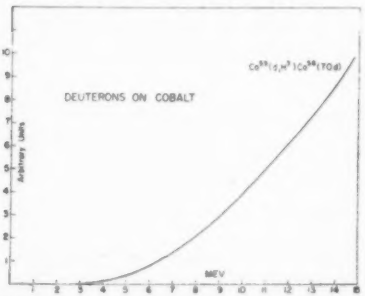

FIG. 3. Excitation curve for $Co^{59}(d,H^3)Co^{58}$ (26). Ordinates are in arbitrary units

The long-lived (3.0 y) isotope of sodium, Na^{22}, is produced by the reaction $Mg^{24}(d,\alpha)Na^{22}$. Figures 4 and 5 give the deuteron excitation function and the thick-target yields for this reaction (27). These figures also give the curves for the simultaneous reaction $Mg^{26}(d,\alpha)Na^{24}$. Long-lived isotopes of vanadium (V^{48}) and manganese (Mn^{54}) are produced by this reaction.

The reaction (d,n) gives rise to several radionuclides that have been used in biological and medical studies. Prepile production of I^{121} and C^{11} were among these. This reaction is probably the first to compete with the (d,p) reaction at intermediate deuteron energies. Figure 6 shows the shape of the excitation curve for this reaction (26).

Many (p,n) reactions have been reported in the literature but the analogous reaction $(d,2n)$ is not so widely

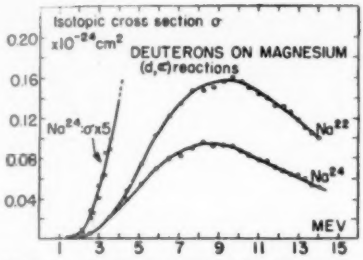

FIG. 4. Excitation curves for deuterons on Mg (27). Curve on left shows points of earlier work of Henderson at lower voltage

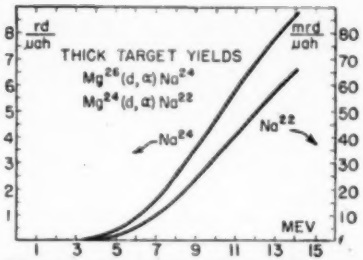

FIG. 5. Thick target yields of Na^{22} and Na^{24} from deuterons on magnesium (27). Ordinates are in rutherfords (10^6 d/s)

recognized. This reaction is endoergic and has a finite, recognizable threshold. Its products nearly always decay by positron emission or orbital electron capture. Very strong sources of positrons (10–15 krd) are available from the reaction $Cr^{52}(d,2n)Mn^{52}$(6.5 d). Table 1 lists several of the longer-lived nuclides obtained in this manner. Figures 1 and 6 show the shape of the deuteron excitation curve for the reaction. In the first of these figures, the $(d,2n)$ reaction appears to start as the major competing reaction with the (d,p) on bromine, and in the second, it seems to compete with the (d,n) reaction at higher energies. Insufficient work has been done with these reactions to establish definitely the exact nature and degree of competition. It is strange that with about 270 stable nuclides and several hundred radionuclides there are almost no nuclides suitable for studying the competing reactions (d,p), (d,n), and $(d,2n)$ in the same compound nucleus. The stacked-foil technique requires a reasonable half-life for the radionuclides produced; and in this group of reactions, at least one of the products is stable, has a half-life too short to use effectively, or other radionuclides produced in the target make activity measurement difficult to interpret.

With the exception of He^{++} for production of radionuclides of the transuranic elements, charged particles other than deuterons are little used for production purposes.

Production Problems

Brief mention should be made of the problems connected with the preparation of targets for bombardment and with chemical processing. The physical requirements for pile targets are generally met with ease. Compounds for these targets should contain a high proportion of the element to be activated. No other element should be present which has a high neutron cross section. This prevents undue radio-

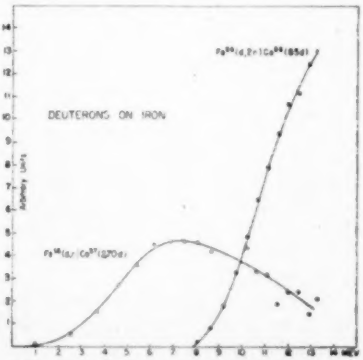

FIG. 6. Excitation curves for (d,n) and (d,2n) on Fe^{56} (26). Ordinates are in arbitrary units

chemical contamination of the product and avoids the danger of upsetting the neutron economy of the pile. The target compound should be moderately stable in high radiation fields and at temperatures of 150°–200° C. The compound should not be corrosive towards its container. Explosive or inflammable compounds naturally cannot be tolerated because of danger to the pile and to people handling the active material.

For bombardment in the outside beam of a cyclotron, the target requirements are much the same as for pile irradiations. If the target material is a poor conductor of heat and the product is volatile, loss may occur due to melting of the target surface.

Cyclotron probe bombardments present a set of conditions that are difficult to meet with any target material. In a high vacuum, the target must dissipate as much as 5–10 kw/cm² if the machine is running at full power. Temperature gradients as high as 1500° C/mm result under these conditions. Even a metal like chromium is melted or volatilized. To decrease the power density on the target, the total power must be reduced to a value suitable for the target being used. This means a loss in operating efficiency of 50–80% but cannot be avoided. Spreading the beam, whose cross section is only ~ 0.5 cm², over a larger area can be accomplished by presenting the target at grazing angles and moving the target in the beam by oscillation or rotation (*28*). Even under the best conditions difficulties are encountered in preparing suitable targets. These problems are discussed by Reid, Weil, and Dunning (*29*).

There are few general statements that can be safely made concerning chemical separation procedures. Since the target may contain anywhere from one to thirty of the 96 chemical elements and will always present a radiation hazard, the processing steps will be dictated by the particular problem at hand.

These unique conditions have stimulated study of unorthodox chemical procedures. Since precipitation reactions are difficult to carry out with high specificity when minute quantities must be handled by remote control, other separation methods are being investigated. One result of these studies has been a spectacular success in achieving separation of the rare earth elements by organic ion exchange resins (*30*). Solvent extraction procedures using selective complexing agents show promise of simplifying the problem of isolating desired radionuclides from complex mixtures of active elements. Electrochemical and high-temperature volatilization also have specific applications.

Much work was done during the war by the Manhattan District and various cyclotron laboratories on processing radioactive mixtures. It is hoped that in the near future some of this work will be published to serve as a guide for people in the field of radiochemistry so the problems connected with the production of radionuclides can be more readily solved.

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Neutron Diffraction and Associated Studies—II

Theory and procedures for the application of neutron diffraction techniques to problems in crystallography, metallurgy, and fundamental nuclear physics are reviewed and interpreted. This is the second of two papers on the subject

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EDITOR'S NOTE: In the first paper published last month, the authors discussed diffraction of neutrons by a single crystal; measurements of resonance absorption; crystal, molecular and nuclear properties; scattering theory; binding, isotope, and spin effects; and transmission measurements with crystalline powders. The numbering of equations and bibliography references in this paper is continued from the first paper.

Debye-Scherrer-Hull Powder Crystal Method

Neutron interference effects in crystalline powders may be studied by direct measurement of the diffracted radiation just as has been widely practiced in the X-ray and electron-diffraction fields. This technique has been developed for neutron-diffraction studies at the Oak Ridge National Laboratory and has been described in some detail in the literature (*34*, *35*). Since the Debye-Scherrer-Hull technique of examining crystalline powders requires a primary beam of monochromatic radiation, a rather severe limitation is placed on the application of this technique to neutron studies. It is quite easy to obtain intense, monochromatic sources of X-rays and electrons but present-day sources of slow neutrons (piles, Ra-Be sources, cyclotrons, etc.) are all of a heterogeneous character so that it is necessary to monochromatize the neutron radiation and this results in a considerable loss in intensity. In spite of this intensity problem, it has been found possible to apply the powder technique with neutrons to a wide variety of problems where high angular resolution in the diffraction pattern is not required.

The arrangement of apparatus used for studying the neutron diffraction pattern of crystalline powders is shown

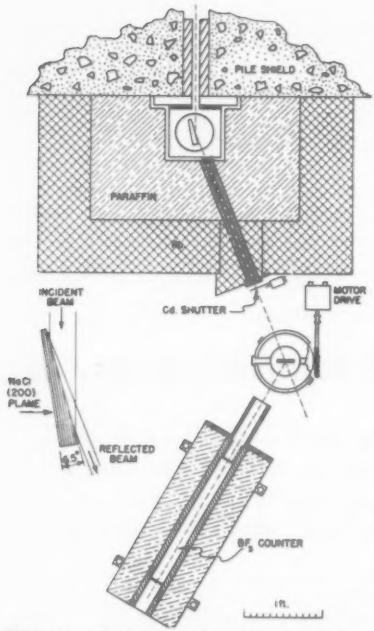

FIG. 9. Schematic diagram of powder diffraction apparatus in use at Oak Ridge. The arrangement of the monochromating crystal (detailed in left center), collimating slits, shielding, the second spectrometer with location of powder specimen, and detecting counter can be seen.

schematically in Fig. 9. A beam of thermal neutrons passes out through a 1-inch square hole in a shield block in the Clinton pile shielding and impinges on a monochromating crystal of NaCl. The (200) planes in this Fankuchen-cut crystal diffract those neutrons having the correct wavelength (1.06 A.U.) for the particular angle of incidence. A further shield of borated paraffin, cadmium, and lead is built up around the monochromating crystal for absorption of the undiffracted neutron and gamma-ray beam. Emergence of the diffracted, monochromatic beam is provided in this shield through a 2-foot-long channel of boron impregnated plastic with an opening ⅜ inch wide and 1 inch high. This beam, of intensity about 2,000,000 neutrons per minute, passes over the center of a spectrometer table where a portion of it suffers further diffraction in the specimen powder. The diffraction pattern is determined by counting the neutron intensity at various angular positions with a high-efficiency BF_3 proportional counter. Alternate readings are taken with a sheet of highly absorbing cadmium in the beam in order to correct for background effects. The equipment has been made to operate automatically through use of a motor drive and a recording counter.

Typical of the diffraction patterns obtained with this equipment are those for diamond and aluminum shown in Fig. 10. The diamond data were taken with an 800-mesh powder whereas the aluminum sample was a ½-inch-thick plate with random orientation of the crystallites brought about by successive heat treatment near the melting point and quenching. Diffraction peaks occur at just those positions dictated by crystal structure and neutron wavelength.

Interpretation of the diffraction pattern in terms of the scattering cross section of the nucleus or nuclei making up the crystal can be made with the aid of the following X-ray powder diffraction equation, in which for the neutron case the polarization factor is omitted,

$$\frac{P_{hkl}}{P_0} = \frac{\lambda^3 l}{4\pi r}\,\frac{h\rho'}{\rho}\,\frac{e^{-\mu h \sec\theta}}{\sin^2 2\theta}\,j_{hkl}N^2F^2_{hkl} \qquad (20)$$

where P_{hkl} is the total power in an (hkl) reflection as measured by a counter slit opening of height l at a distance r from the sample, P_0 the primary beam power, λ the neutron wavelength, ρ' the apparent density of the powder, ρ the density of the solid crystal, $e^{-\mu h}$ the transmission of the specimen of thickness h, θ the glancing or Bragg angle of reflection, j_{hkl} the multiplicity value, N the number of unit cells per cm³ of crystal and F_{hkl} the Bragg scattering amplitude per unit solid angle per unit

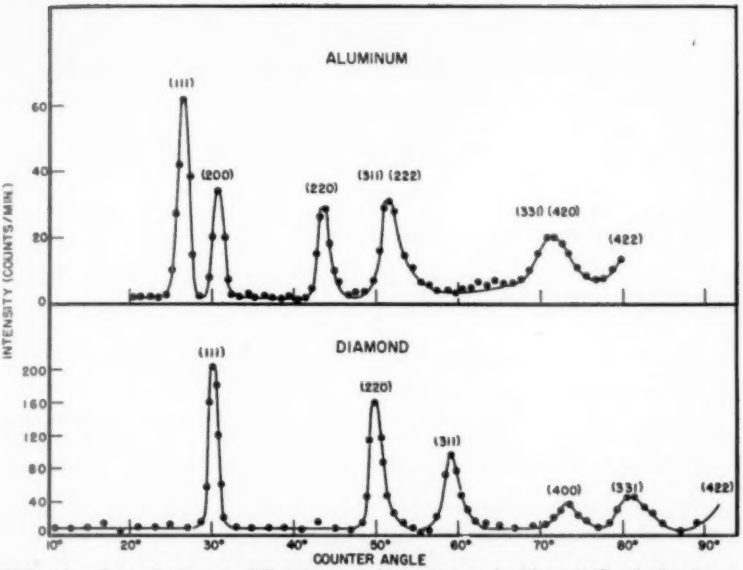

FIG. 10. Typical neutron diffraction powder patterns for diamond and aluminum. The diffuse scattering in the diamond pattern is caused by multiple scattering in the specimen whereas the diffuse scattering in the pattern for aluminum is true thermal scattering, especially at the larger angles.

cell of the crystal. F_{hkl} corresponds to the usual crystal structure factor in X-rays, except that here it is a true amplitude with the dimension of length, and is a simple function of the nuclear scattering amplitudes present in the crystal. For instance

$$F_{111}\ \text{(diamond)} = 4\sqrt{2}\, f_c \qquad (21)$$

where f_c is the effective nuclear scattering amplitude of carbon and the numerical factor is determined by a suitable summation (*36*) over all atoms in the unit cell. For polyatomic crystals, F_{hkl} will in general contain all of the various nuclear scattering amplitudes.

Since the atoms, and hence the nuclei, are in thermal agitation in the crystal, the effective scattering amplitude will be somewhat smaller than the true nuclear scattering amplitude. This can be corrected for by application of the Debye-Waller temperature factor so that

$$f_T = f_0 e^{-W} \qquad (22)$$

with

$$W = \frac{6h^2}{mk\,\Theta}\left(\frac{\phi(x)}{x} + \frac{1}{4}\right)\frac{\sin^2\theta}{\lambda^2}, \quad x = \frac{\Theta}{T} \qquad (23)$$

where h is Planck's constant, m is mass of atom, k is Boltzmann's constant, Θ is the Debye characteristic temperature and $\phi(x)$ is the Debye function. Weinstock (*24*) has shown the validity of the application of this correction factor to the case of Bragg scattering of neutrons.

The diamond pattern in Fig. 10 has been used to standardize the powder diffraction patterns in terms of absolute scattering cross sections. Measurements of the transmission of diamond cells indicate that the total scattering cross section for a bound carbon nucleus is 5.2 barns. Since carbon is mono-

TABLE 1

Results of Powder Diffraction Analysis of Scattering Data for Aluminum

hkl	P_{hkl} $\left(\frac{integrated}{counts/min}\right)$	$f_T(10^{-12}\ cm)$		$f_0(10^{-12}\ cm)$	σ_{coh} (*barns*)
111	82.0	0.317		0.330	1.38
200	47.5	0.321		0.338	1.43
220	20.3	0.301		0.334	1.40
311 222	91.2	0.302		0.350	1.54
331 420	73.9	0.268		0.345	1.50
			averages	0.340	1.46

isotopic to within one percent and since this isotope possesses no nuclear spin, it would be expected that the total scattering cross section would equal the coherent scattering cross section. This is borne out by measurements of the diffuse scattering by the crystal. After corrections were made for the effects of multiple scattering by the sample, the diffuse scattering was found to be zero within the accuracy of the experiment. Accordingly, all other patterns are compared to that of diamond, with 5.2 barns being taken as the coherent scattering cross section for carbon. On this basis the coherent scattering cross section for aluminum has been evaluated from the integrated intensities represented in the pattern of Fig. 10.

The data for the lines in this pattern are listed in Table 1. The third column gives the measured scattering amplitudes, f_T, which decrease with sin θ because of effects of thermal agitation of the atoms in the crystal. Multiplying the values of f_T by the Debye-Waller temperature factor gives the values of $f_0 = f_T e^W$ listed in the fourth column. The values of f_0 are constant within experimental error and their average represents the true coherent scattering amplitude of the aluminum nuclei in the crystal. The observed coherent scattering cross section $\sigma_{coh} = 4\pi f_0^2 = 1.46$ barns is found to agree closely with the total scattering cross section of 1.48 barns for bound aluminum nuclei as taken from literature data. This close agreement between the total and coherent scattering cross section shows that the scattering by aluminum exhibits little or no spin dependence.

Diamond and aluminum represent the simplest of cases in the diffraction of neutrons by crystals since in each case only one element is involved and this consists of a single isotope to which a single scattering amplitude can be assigned since there is little or no dependence on spin orientation. With monoatomic crystals, measurement of the absolute intensity of a single diffraction peak, plus a knowledge of the characteristic temperature of the crystal, suffices for the determination of the coherent cross section. With polyatomic crystals, two or more peaks must be measured to obtain cross sections for the individual atoms, and even then a definite assignment to the individual nuclei can be made only by measurements with a given element in several compounds. For X-rays, the assignment of amplitude to a given scattering center is nearly always unambiguous since atomic scattering factors are

TABLE 2

Results of Diffraction Analysis for Various Sodium-Containing Crystals

Crystal	$f_0(10^{-12}$ *cm*) Na	X	σ_{coh} (*barns*) Na	X	σ_{total} (*barns*)
Na	0.345	—	1.50	—	3.7 (Na)
NaBr	0.335	0.65	1.41	5.3	7.5 (Br)
NaCl	0.358	1.01	1.61	12.8	15 (Cl)
NaF	0.348	0.65	1.52	5.2	4.5 (F)

known to increase with atomic number.

As an example of a problem involving polyatomic crystals with spin and isotope effects, the scattering by a number of sodium-containing crystals will be considered. Powder diffraction measurements have been made on Na, NaBr, NaF, and NaCl from which the coherent scattering amplitudes for sodium and the other elements in the crystal have been determined. For NaBr, NaCl and NaF, the (111) and (200) reflections suffice to determine the amplitudes of the two elements in the crystal, and for sodium only one reflection (111) is required. The results of the measurements of these sodium-containing crystals are tabulated in Table 2. Scattering amplitudes for sodium and the other atomic constituents, corresponding coherent scattering cross sections, and total scattering cross section for bound nuclei are given. It will be noticed from the table that the coherent cross sections for Br and Cl are somewhat smaller than the total scattering cross sections for these elements. Since each of these elements contains two isotopes, it is likely that this difference is attributable to an isotopic effect. In the case of fluorine, which is monoisotopic, it will be noticed that the measured coherent cross section is slightly larger than the total cross section. It is, of course, not possible for this actually to be the case and hence the difference must be due to experimental error in one of the two measurements.

The observed cross section values for sodium, as determined from the four crystals, are seen to be in reasonably good agreement, giving an average value of $\sigma_{coh} = 1.51$ barns. This value of the coherent cross section is considerably lower than the total scattering cross section of 3.7 barns for bound nuclei and since sodium is monoisotopic the scattering by sodium must be spin dependent. From the above values of the coherent and total scattering cross sections it is possible to determine the separate amplitudes for the parallel and antiparallel spin states. Solution of Eqs. 14 (modified to the case of bound centers) and 15 for $f_{i+\frac{1}{2}}$ and $f_{i-\frac{1}{2}}$ gives

$$f_{i+\frac{1}{2}} = f_0 \pm \sqrt{\frac{\sigma_b - \sigma_{coh}}{4}} \times \frac{i}{i+1}$$

and

$$f_{i-\frac{1}{2}} = f_0 \mp \sqrt{\frac{\sigma_b - \sigma_{coh}}{4\pi}} \times \frac{i+1}{i}$$

from which these amplitudes can be determined with the additional knowledge that the spin of sodium is $\frac{3}{2}$. Since these equations are quadratic they lead to the two sets of values, $f_{i+\frac{1}{2}} = +0.88$ or -0.20 and $f_{i-\frac{1}{2}} = +0.02$ or $+0.67$, respectively (all $\times 10^{-12}$ cm). There is nothing in the present experiment which would permit a choice to be made between these two sets of values.

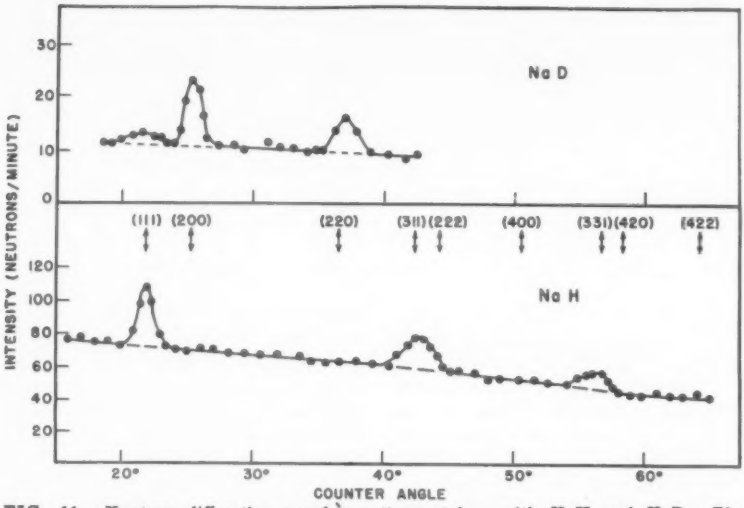

FIG. 11. Neutron diffraction powder patterns taken with NaH and NaD. The pronounced differences in the two patterns are caused by differences in neutron scattering by hydrogen and deuterium. It is to be noted that the diffuse scattering is much stronger for NaH than for NaD.

Applications of Neutron Diffraction to Crystallographic Studies

One of the most attractive potential applications of neutron diffraction is the determination of the location of hydrogen atoms in hydrogeneous crystals. Such determinations have been possible with X-rays only in rare cases because of the extremely low scattering power of the hydrogen center for X-rays. With neutrons, however, the coherent scattering cross section for hydrogen or deuterium turns out to be quite comparable with that of other nuclei. Aside from the crystallographic interest in hydrogen scattering, the scattering of neutrons by protons or deuterons is of great importance from the nuclear physics point of view since these simplest of nuclear reactions can be treated theoretically for comparison with experiment.

As an introduction to studies of this nature, the diffraction of neutrons by the crystalline powders NaH and NaD has been studied at Oak Ridge by Shull, Wollan, Morton and Davidson (*35*), using the powder spectrometer already described. X-ray diffraction studies of NaH have shown the sodium centers to lie at face-centered cubic positions (as in NaCl) but because of the low scattering power of the single electron in the hydrogen atom, the positions of the latter atoms cannot be established. By analogy, however, it is suspected that hydrogen atoms occupy positions similar to those of chlorine atoms in NaCl.

The neutron diffraction patterns for NaH and NaD are shown in Fig. 11. Pronounced peaks having odd indices and weak or absent peaks having even indices are to be noted for NaH in contrast to the prominent (200) peak and weaker (111) peak in NaD. This shows immediately that hydrogen and deuterium are contributing to the diffraction pattern since the pattern caused by sodium only (as in the X-ray

diffraction case) would possess a (200) peak about half as strong as the (111) peak. It furthermore is suggested that hydrogen and deuterium are scattering neutrons with a phase reversal relative to each other but, as yet, nothing can be said about the relative phase of scattering of sodium compared with the other two centers because of the possible uncertainty in the crystal structure.

Fortunately it is rather easy to establish the correctness of the suspected NaCl-type structure from a study of the outer peak intensities for NaH. Among the several structures of face-centered cubic type which are known to exist, there are only two (B-1 type, NaCl and B-3 type, ZnS) possible structures for NaH. Other structures are ruled out because of unit cell size. A comparison of the measured intensities with the squares of the structure factors (approximately proportional to the intensities) calculated for these two structures is given in Table 3. It is seen that either the ZnS structure with a common sign for f_{Na} and f_H or the NaCl structure with opposite signs for the amplitudes would satisfy the experimental data for the (111) and (200) peaks, but the relative intensities of the (220), (311), (420), and (331) eliminate the ZnS structure from consideration. For instance the (220) peak of the ZnS structure should be about as strong as the (111) and (311) peaks and this is in contradiction to the experimental data. Similar considerations with respect to the (420) and (331) peaks lead to the same conclusion. Thus it is established that *sodium hydride possesses the same type of structure as NaCl and that sodium and hydrogen are scattering neutrons with opposite phase. It also follows that sodium and deuterium are scattering neutrons with common phase.*

The pattern intensities shown in Fig. 11 can be used for evaluating the scattering amplitudes and hence the

TABLE 3

Experimental Intensities and Values Calculated for Possible Structures of NaH

Diffraction Peak	*Experimental Intensity*	*Calculated for* ZnS *Structure*	*Calculated for* NaCl *Structure*
(111)	37	$f^2_{Na} + f^2_H$	$(f_{Na} - f_H)^2$
(200)	<2	$(f_{Na} - f_H)^2$	$(f_{Na} + f_H)^2$
(220)	0	$(f_{Na} + f_H)^2$	$(f_{Na} + f_H)^2$
(311)	25	$f^2_{Na} + f^2_H$	$(f_{Na} - f_H)^2$
(222)	0	$(f_{Na} - f_H)^2$	$(f_{Na} + f_H)^2$
(400)	<5	$(f_{Na} + f_H)^2$	$(f_{Na} + f_H)^2$
(331)	15	$f^2_{Na} + f^2_H$	$(f_{Na} - f_H)^2$
(420)	0	$(f_{Na} + f_H)^2$	$(f_{Na} + f_H)^2$

coherent scattering cross sections of the various nuclei. For this purpose the scattering amplitude for sodium has been taken from earlier described measurements, and after suitable temperature corrections have been applied for thermal motion of the centers, the coherent scattering cross sections for hydrogen and deuterium are evaluated as

$$\sigma^H_{coh} = 2.0 \pm 0.3 \text{ barns}$$
$$\sigma^D_{coh} = 4.1 \pm 1.2 \text{ barns} \qquad (24)$$

The estimated errors in these cross sections are derived from the uncertainties in the intensity measurements and in the quantitative composition of the hydride and deuteride preparations. Thus it is seen that the coherent scattering or diffractive power of hydrogen and deuterium is conveniently near that of other nuclei and that crystallographic studies of hydrogen positions in other crystals should be readily possible.

The above coherent scattering cross section for hydrogen of 2.0 barns is very much smaller than the total bound scattering cross section of 80 barns and this is a result of the large spin dependence of neutron scattering by

protons. Pronounced spin dependence of scattering leads to a large incoherent scattering (see earlier section) and this is to be noted in the diffuse background of the NaH pattern in Fig. 11. In contrast, the diffuse scattering in the NaD pattern is seen to be much less pronounced because the coherent cross section of 4.1 barns for deuterium is more nearly comparable to the total scattering cross section of 7.0 barns for bound deuterium centers.

The presence of the large diffuse scattering obtained with hydrogen-containing crystals makes difficult the measurement of the relatively small coherent features of the pattern and hence it would seem that crystallographic studies could be more easily performed with deuterated compounds for which the structure is, of course, identical to that of the hydrogeneous material. Evidence supporting this conjecture has been obtained in unpublished work at Oak Ridge on the structure of water and ice. The powder diffraction pattern for ice was obtained much more easily with heavy ice than with ordinary ice.

There exist many other crystallographic problems similar to that of the location of hydrogen positions which appear also to be promising applications of neutron diffraction techniques. For structures where one type of atom has a much larger scattering power than the other, complete crystallographic analysis by X-ray or electron diffraction becomes very difficult. This is the case, for instance, for PbO in which the scattering power for lead is so much larger than that for oxygen that little information on the oxygen positions can be obtained. With neutrons, however, the scattering cross sections are not nearly so widely separated so that suitable neutron diffraction studies should locate the oxygen centers.

Neutron diffraction or scattering techniques have been applied recently to two problems of metallurgical interest. Sidhu (*37*) has reported studies on the solubility of WC in TiC from measurements of the transmission cross section for a series of solutions and mechanical mixtures. He was able to show the presence and extent of solubility of the one constituent in the other from the neutron measurements. Very recently, unpublished work performed at Oak Ridge on FeCo alloys has shown the presence of superstructure in certain heat-treated samples of this system. Powder diffraction patterns were obtained, and for certain preparations these showed the presence of extra superstructure diffraction lines characteristic of an ordered array of the two atoms, in contrast to the usual substitutional solution of one atom in the lattice of the other. The FeCo system is a very difficult one to study with X-rays since the atomic structure factors for the two atoms are so nearly the same.

Scattering of Neutrons by Gas Molecules

The theoretical treatment of the scattering of neutrons by gases is more complicated than the corresponding X-ray problem because with neutrons there may be spin and binding effects and also the inelastic processes have a more pronounced influence on the scattering cross section.

A theory for the scattering of neutrons by hydrogen has been developed by Schwinger and Teller for low rotational states of the molecule. Since with hydrogen the scattering is strongly spin-dependent, the ortho and para forms, in which the nuclear spins for the two atoms in the molecule are, respectively, parallel and antiparallel, have markedly different scattering cross sections. Since the hydrogen problem is of importance in connection with the theory of the deuteron and neutron-proton interaction, it will be discussed in a later section.

TABLE 4

Scattering Cross Sections for Various Room-Temperature Gases with Very Slow Neutrons $\lambda = 5.1$ A (from Fermi and Marshall)

Molecule	σ (*observed*)	*Sum of the total cross sections of constituent atoms*	σ *calculated from classical interference theory*
CO_2	24.5 barns	13.0 barns	24.8 or 4.1 for opposite phase
N_2O	57.8	34	55 or 41 for opposite phase
O_2	16.2	8.2	13.2
N_2	47.4	30	44.4
CF_4	41.5	21	38 or 7.5 for opposite phase

The neutron scattering cross sections for some gas molecules have been investigated experimentally by Fermi and Marshall (*29*). They measured the transmission of very-low-energy neutrons through a tube 365 cm long, containing the gases at room temperature and at pressures up to 2 atmospheres. The low-energy neutrons were obtained by filtration through a BeO column which gives the spectrum shown in Fig. 8 as previously described. For neutrons of wavelength short compared to the interatomic distances, the molecular scattering cross section is the sum of the individual atomic cross sections. With the long wavelengths (~ 5 A.U.) used in these experiments, interference effects tend to increase the observed cross sections. It would be difficult to calculate accurately the intensity to be expected for the scattering of neutrons by molecules such as N_2 and O_2, but approximate results can be obtained by considering the problem to be identical to the corresponding X-ray problem.

Table 4 shows the results of Fermi and Marshall for a number of gases. The second column gives the observed scattering cross sections, the third column gives the sum of the total cross sections of the constituent atoms and the fourth column gives the cross sections as calculated on the basis of classical interference theory for the case of equal and of opposite phase for the atoms in the molecule. It will be noticed that the classical interference theory gives results which agree well with the experimental data when equal phases are assumed for the atoms in the molecules.

Neutron-Proton Scattering

The scattering of neutrons by protons, being a two-body problem, is amenable to quite exact theoretical analysis. An early treatment of this problem was made by Wigner and by Bethe and Peierls on the assumption of a short-range neutron-proton interaction and, together with a later modification to a square-well potential by Bethe and Bacher, the following expression for the scattering cross section was obtained

$$\sigma = \frac{4\pi\hbar^2}{M} \frac{1 + \alpha_0 r_0}{|E_0| + \frac{1}{2}E} \qquad (25)$$

where $|E_0|$ is the binding energy of the deuteron, and E is the neutron energy measured in the system in which the proton is at rest, r_0 is the range of the potential well and $\alpha_0 = (M \cdot |E_0|/\hbar^2)^{1/2}$.

With the known values of E_0 and M and with a reasonable value of r_0 (2×10^{-13} cm), this expression predicts a cross section of 3.5 barns for slow neutrons as against the measured value of about 20 barns. It can be

shown that the theory is insensitive to the shape of the assumed potential well. To explain this marked disagreement, Wigner suggested that the neutron-proton interaction in the singlet state (antiparallel neutron and proton spins) differs from that in the triplet state (parallel spins). With this assumption he determined the scattering cross section to be

$$\sigma = \frac{4\pi\hbar^2}{M}$$

$$\left[\frac{3}{4}\frac{1+\alpha r_0}{|E_0|+\frac{1}{2}E}+\frac{1}{4}\frac{1+\alpha_1 r_0}{|E_1|+\frac{1}{2}E}\right] \quad (26)$$

where $|E_1|$ is the binding energy in the singlet state and

$$\alpha_1 = \pm(M\cdot|E_1|/\hbar^2)^{\frac{1}{2}}$$

the plus or minus sign being obtained according to whether the singlet state is real or virtual. A check of this expression can be obtained by a measurement of $|E_1|$. Assuming the theory to be correct, $|E_1|$ can be tentatively evaluated from the known value of σ. Schwinger and Teller calculated $|E_1|$ = 142,000 ev for a real level and $|E_1|$ = 114,000 ev for a virtual level using the then-available data for σ.

It became important then to devise measurements from which E_1 could be determined directly. Fermi showed that a measurement of the capture cross section of neutrons by protons would permit the correct choice to be made between these two possibilities and such measurements have indicated the existence of a virtual singlet state for the deuteron. Schwinger and Teller (*7*) pointed out that a direct test of Wigner's hypothesis of a spin dependent interaction between neutrons and protons could be obtained from experiments on the scattering of slow neutrons by ortho- and parahydrogen. Recently the diffraction of neutrons by hydrogen-containing crystals has furnished an additional method for studying the spin dependence of scattering by protons.

Neutron-Proton Scattering Amplitudes from Ortho- and Parahydrogen Scattering and from Neutron Diffraction. Since in orthohydrogen the spins of the two protons are parallel and in parahydrogen they are antiparallel, the interference effects between the neutron waves scattered by the two protons will be different if the scattering amplitude depends on the relative orientation of the spins of the neutron and proton. The theoretical determination of the intensity of scattering by molecules is complicated by the fact that in addition to elastic scattering there will be inelastic processes in which rotation and vibration levels will be excited. With suitable conditions of neutron wavelength and temperature and physical state of the scattering molecules, however, by the theory as developed by Schwinger and Teller, the experimental data can be interpreted quantitatively in terms of the scattering amplitudes characteristic of the two spin states.

Early measurements by Halpern, Eastermann, Simpson and Stern (*8*), and by Brickwedde, Dunning, Hoge and Manley (*8*) of the scattering of slow neutrons by various concentrations of ortho- and parahydrogen in the liquid form showed the scattering cross section for orthohydrogen to be several times larger than for parahydrogen. These results were sufficient to show that the neutron-proton interaction is spin-dependent and that the singlet state of the deuteron is virtual. Since these experiments were performed with Ra-Be neutrons slowed down in cold paraffin, there were neutrons of sufficient energy to excite para to ortho transitions; hence the elastic para cross section could not be measured. The fact that the scattering was measured on a liquid may also have introduced intermolecular effects which could not be accurately interpreted. Alvarez and Pitzer made measurements with hydro-

TABLE 5

Scattering Data and Range of Forces Resulting from Ortho-Para-Hydrogen Scattering

	Observed		*Calculated*			
Neutron Energy (10^{-3} *ev*)	σ_{ortho} (*barns*)	σ_{para} (*barns*)	a_1 (10^{-12} *cm*)	a_0 (10^{-12} *cm*)	σ_f (*barns*)	r_0 (10^{-12} *cm*)
0.867	145	4.98	+0.505	−2.32	19.3	1.33
1.250	133	4.40	+0.510	−2.33	19.6	1.43
1.463	128	4.19	+0.518	−2.34	19.8	1.58
1.723	124	3.97	+0.520	−2.34	19.8	1.61
1.830	122	3.90	+0.522	−2.35	19.9	1.65
2.386	116	3.58	+0.526	−2.35	19.9	1.72

gen gas as scatterer and they used the modulated cyclotron method to obtain a monoenergetic beam of very slow neutrons. Although the conditions under which this latter experiment was performed were very satisfactory, there seemed to be some question regarding the values of the ortho- and parahydrogen scattering cross sections obtained in this experiment.

This experiment was recently repeated (*38*) by a group working at Los Alamos under what seem to be very carefully controlled conditions. As in the previous experiment, transmission measurements were made on samples of ortho- and parahydrogen gas maintained at a temperature of about 20° K. One sample consisted of 99.9% parahydrogen and the other sample consisted of normal hydrogen which has a concentration 75% ortho- and 25% parahydrogen at room temperature, correction being made for the change in concentration at the low temperatures at which the measurements were made. The cross sections were measured as a function of neutron energy over the range 0.0008 ev to 0.0025 ev.

With the slow neutrons used in this experiment and with the low temperature (20° K) at which the hydrogen gas was maintained, the scattering cross sections are given by

$$\sigma_{para} = 6.47\,(3a_1 + a_0)^2$$

and

$$\sigma_{ortho} = 6.29\,[(3a_1 + a_0)^2 + 2(a_0 - a_1)^2] + 1.45(a_0 - a_1)^2 \qquad (27)$$

where the last term in the ortho cross section represents the inelastic scattering involving the conversion of ortho to para. The numerical coefficients were calculated from the theory (*39*) for the conditions of the experiment.

The experimental data of the Los Alamos investigation are shown in Table 5. This table gives the neutron energy, observed ortho- and parahydrogen scattering cross sections, triplet and singlet scattering amplitudes, total scattering cross section for free protons as calculated from the observed amplitudes, and range, r_0, of the neutron-proton triplet interaction.

The range of the square-well triplet interaction has been calculated from the triplet amplitude a_1 with the aid of a theoretical calculation by Breit and Keitel. The average value of this range, $r_0 = 1.54 \times 10^{-13}$ cm, is considerably smaller than the range for the proton-proton singlet interaction which has been found to be 2.8×10^{-13} cm and indicates a real difference between these two types of nuclear interaction. Information on the amplitudes of spin scattering can also be obtained (*35*) from the value of the coherent scattering cross section for hydrogen as determined by the diffraction analysis of NaH. As discussed in an earlier section, this cross section is given by

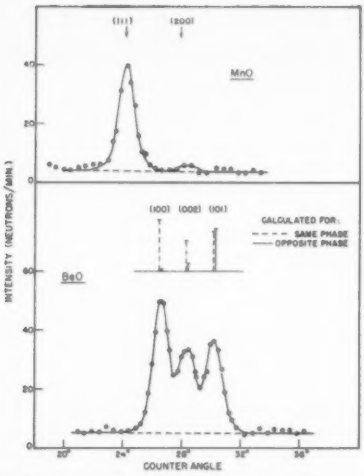

FIG. 12. Intensity of Bragg reflections from MnO and BeO. The small (200) and large (111) reflections from MnO show that Mn and O scatter with opposite phase. Also a comparison of the measured and the calculated intensities for BeO with the same and opposite scattering phase for Be and O shows that these centers are scattering with the same phase in agreement with the conclusions from Fig. 7.

$$\sigma^{H}_{coh} = \pi(3a_1 + a_0)^2 \qquad (28)$$

Combining this with the value of the free proton scattering cross section given by

$$\sigma^{H}_{free} = 4\pi(\tfrac{3}{4}a_1^2 + \tfrac{1}{4}a_0^2) \qquad (29)$$

permits evaluation of the scattering amplitudes. For this purpose, $\sigma^{H}_{coh} = 2.0$ barns from Eq. 24, and the direct measurement of $\sigma^{H}_{free} = 20$ barns by Melkonian (*40*), have been used to give the result

$$\begin{aligned} a_1 &= +0.520 \cdot 10^{-12}\ \text{cm} \\ a_0 &= -2.35 \cdot 10^{-12}\ \text{cm} \end{aligned} \qquad (30)$$

These values are in good agreement with those resulting from the ortho- and parahydrogen experiment and lead to a triplet range

$$r_0 = 1.6 \pm 0.2 \cdot 10^{-13}\ \text{cm} \qquad (31)$$

which is also in good accord with the result obtained from the ortho-parahydrogen scattering experiments.

Phase of Scattering

The scattering of neutrons by nuclei, as we have seen, is expressible in terms of the shift in phase of the total wave for neutron and nucleus, relative to the incident wave in the absence of a scattering center. These phase shifts are in general very small and the amplitude of scattering is then proportional to the magnitude of this phase shift as shown by Eq. 8. Small positive and negative phase shifts correspond to scattering amplitudes of opposite sign and hence to scattered waves which are 180° out of phase with each other.

The relative phase of the scattered waves from two atomic species can be determined by studying the interference effects when these atoms are together in a crystal or molecule. When more than one type of atom is present in the crystal, the structure factors F_{hkl} and hence the diffraction line intensities will show a pronounced dependence upon the relative phase of scattering of the different atomic species. Thus the diffraction pattern affords a convenient means for such determinations.

By studying the intensity of diffraction from different planes in a variety of single crystals, Fermi and Marshall were able to deduce the relative phases of scattering for approximately fifteen elements. Most of these turned out to be of the same scattering phase but two elements, namely Li and Mn, were found to scatter with opposite phase. Similar studies with a large number of crystalline powders have been made by Sidhu and Weber using the transmission method and by Shull and Wollan using the diffraction method, so that the number of elements possessing known scattering phase now totals around 30.

Typical of the diffraction patterns obtained are those shown in Fig. 12 for MnO and BeO. A comparison of the

experimental intensities with those calculated for the same and reversed phases of scattering by the elements in the crystal shows conclusively that Be and O possess the same phase of scattering in contrast to the reversed phase shown by Mn. Similar conclusions on Be and O were arrived at from the transmission data of Fermi, Sturm and Sachs, shown in Fig. 7. The experiments discussed in a previous section on the scattering by gas molecules also provided information on the relative phase of scattering of the atoms involved.

Although the relative phase of scattering can be readily determined in interference experiments, such experiments do not determine directly whether the scattered wave is 180° out of phase with the incident wave or whether it is in phase, i.e., the absolute value of the phase shift. As has already been mentioned in a previous paragraph, negative phase shifts which we take as corresponding to positive scattering amplitudes, that is, to scattered waves which are 180° out of phase with the incident wave, are much more probable than negative scattering amplitudes in which the scattered waves are in phase with the incident wave. This in itself should permit one to assign the proper sign to the scattering amplitude for many of the elements which have been paired in the above-mentioned scattering experiments. From some experiments, however, it is possible to obtain a direct determination of the sign of the scattering amplitude.

This is true in the case of measurements of the total reflection of neutrons. The index of refraction for neutrons is given by the equation $n = 1 - \lambda^2 Nf/2\pi$ where f is the scattering amplitude taken with a proper sign, N is the density of atoms in the reflecting surface, and λ is the wavelength. It is seen from this equation that n will be less than 1; hence the surface of the material will give rise to total reflection if the scattering amplitude is positive. In the case of neutrons, as with X-rays, $n - 1$ is a small quantity of the order of magnitude of 10^{-6} and the critical angle for reflection will be of the order of 10 min.

TABLE 6

Sign of Scattering Amplitudes

Positive Scattering	*Negative Scattering*
D, Be, C, N, O F, Na, Mg, Al, S Cl, K, Ca, Fe, Co Ni, Cu, Zn, Ge, Br Sr, I, Cs, Ba, Au Tl, Pb	H, Li, Ti, Mn

Fermi, Zinn, and Marshall (*41*) have made experiments in which they measured the critical angle of reflection of a beam of neutrons from mirror surfaces of a number of materials. In these experiments they were able to show that Be, Cu, Zn, Ni, Fe and C scatter neutrons with positive scattering amplitudes. So far we have considered only the effective scattering phase for elements as a whole. If an element consists of more than one isotope, the scattering amplitudes for the two isotopes may be of opposite sign. In the case of Li, it has been shown in recent unpublished measurements by Shull and Wollan on the diffraction by the separated Li^7 isotope that the negative scattering amplitude observed with normal Li is to be associated with the scattering by Li^7 and that Li^6 scatters with positive amplitude.

In cases where the scattering is found to be spin-dependent, there will be two scattering amplitudes associated with a given nucleus, one for parallel and one for antiparallel spin orientation of neutron and nucleus. In this case the sign of the scattering amplitude may be different for the two spin states. This situation is exemplified in the

scattering by hydrogen, in which case it has been possible to determine both the magnitude and sign of the scattering amplitudes for the parallel (triplet) and antiparallel (singlet) spin states. In this case the negative scattering amplitude for the singlet state predominates to give hydrogen an effective negative scattering amplitude.

A summary of the effective scattering phases for elements on which measurements have been made to date is given in Table 6.

Laue Photography of Neutron Diffraction

In studies of crystal and molecular structures by means of X-rays and electrons, the photographic technique has played a very important role. This technique may also prove useful in connection with various neutron diffraction problems. The detection of neutrons, however, is a very different problem from the detection of X-rays and electrons which produce ionization in any matter through which they traverse. Neutrons, being non-ionizing in themselves, are usually detected by the ionizing radiation emitted from a nucleus which has captured the neutron. The most common form of detector for slow neutrons is a boron-filled proportional counter in which the neutrons are captured in the reaction $B^{10} + n^1 \rightarrow Li^7 + He^4$ and the ionization is produced by the emitted alpha particle and the recoiling Li^7 nucleus. Photographic film containing boron, lithium or nitrogen will be sensitive to slow neutrons due to the emission of the ionizing particles arising in the corresponding reactions. The sensitivity of films impregnated with these elements will depend on the neutron cross section for the element and on the amount of the element which the emulsion can be made to hold.

An alternative method of obtaining photographic effects with neutrons is to place the boron or other neutron-sensitive material in contact with the film, in which case the ionizing particles that escape from sensitive layer will impinge on the film and thus produce photographic darkening. The sensitivity of this photographic method of detection would, of course, be much greater if the tracks of each individual particle were observed through a microscope since many more particle tracks per unit area are required to produce photographic darkening than are useful for counting purposes. The counting technique is so laborious, however, that it does not appear at all feasible for the applications at hand. It appears that the most sensitive and convenient method now available for obtaining photographic darkening on a film by neutrons is to utilize the beta rays resulting from artificial radioactivity induced by neutron capture. A sheet of indium placed adjacent to ordinary X-ray film serves as a sensitive screen to record incident neutron intensities.

Although the present neutron intensities available from reactors are adequate for many neutron diffraction techniques, they are still considerably lower than those available from modern, high-power X-ray sources. This fact limits to a great extent the possible applications of the photographic technique. Since the neutron radiation from a reactor is heterogeneous and contains a thermal distribution of energy or wavelength, the photographic method would seem most easily applied to the Laue technique. In the Laue method, a continuous distribution of wavelength is required and the examination is performed with a single crystal. The various planes of diffracting centers in the crystal have definite orientations with respect to the incident beam and diffraction occurs only for those neutrons possessing the correct wavelength required by the Bragg law.

By using indium as a sensitizing screen next to X-ray film as described

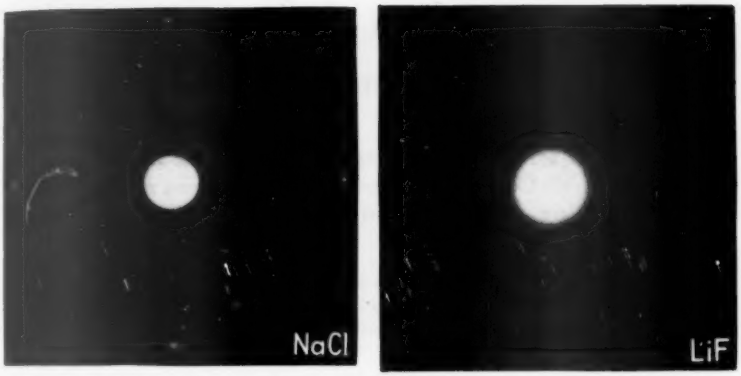

FIG. 13. Neutron diffraction Laue photographs taken with single crystals of NaCl and LiF. The fact that the intensities of the inner and outer spots in the LiF pattern are reversed with respect to the spots in the NaCl pattern is a consequence of the negative scattering amplitude of Li compared to the positive amplitude for F, Na and Cl.

above, Wollan, Shull and Marney (*42*) recently obtained Laue photographs with single crystals irradiated by a thermal neutron beam from the Oak Ridge reactor. A well-collimated beam ¼ inch in diameter was used with a crystal-to-film distance of 6.4 cm, in which case exposures of about 10 hours were required to obtain a picture. Figure 13 shows patterns obtained for single crystals of NaCl and LiF for which the neutron beam was passed parallel to one of the cube edges. The white spot in the center of the patterns corresponds to a hole in the backing plate of boron-impregnated plastic, with the irradiation of this central region being produced by backscattering from the film holder. The larger circular edge corresponds to a 1½ inch hole cut in the indium sheet.

Since NaCl and LiF possess identical face-centered cubic structures, Laue spots should be located at the same positions on the two patterns but should show some intensity differences because of the different scattering power of the various atoms. Indexing of the Laue spots by the usual gnomonic projection shows that the outer series of spots corresponds to reflections from planes with indices (402), (422) and their permutations, while the inner group of spots represents reflections from planes (311) and permutations. Other spots such as the (442) and (513) are visible on the original negative but are too weak to show on the reproduction. The even-indexed spots in the NaCl pattern are somewhat more intense than those with odd indices, whereas the reverse is true for the LiF pattern. This is a consequence of the phase reversal of neutron scattering from a lithium nucleus.

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Effect of High-Voltage X-Rays and Cathode Rays on Vitamins (Niacin)*

Results of extensive tests using three million-volt radiations from Van de Graaff-type generator to irradiate pure solutions of niacin are presented in first report published on subject

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A NUMBER of investigators have shown that X-rays and cathode rays have a lethal effect on microorganisms (*18–20, 24, 30*). The possibility of utilizing X-ray exposures in sterilizing foods was demonstrated by Proctor, Van de Graaff, and Fram in 1943 (*22*). Dunn *et al.* (*7*) found that both X-rays and cathode rays have a lethal effect on a number of bacteria, yeasts, and molds, and they successfully pasteurized milk with the use of cathode rays. If such radiations have potentialities as a means of processing foods, information is essential concerning their effects on vitamins, enzymes, and other nutrients in foodstuffs.

The present paper deals with the effects of hard X-rays and cathode rays produced at three million volts on pure solutions of niacin. The authors know of only three published reports on the quantitative effects of X-rays on vitamins, namely, on ascorbic acid (*1*), *p*-aminobenzoic acid (*25*), and thiamine (*6*). In these instances the investigators used soft X-rays produced at relatively low voltages, 200 kv or less. No published reports have been found on the effect of X-rays on niacin. As compared with soft radiations, the advantage of using high-voltage X-rays is their greater degree of penetration. In other words, as the voltage increases, the wavelength diminishes and the depth to which roentgen rays penetrate matter increases because of the progressive reduction of both the photoelectric and the Compton scattering processes of absorption.

Niacin is a relatively stable molecule. The pyridine ring is noted for its aromaticity and is resistant to oxidation. Information on the oxidation of either pyridine or niacin is lacking. It has been clearly indicated by other investigators, however, that the action of X-rays and cathode rays on other compounds appears to be one of oxidation (*12–14*) or of reduction (*2, 3, 11*), depending on the oxidation-reduction potential. Warburg has shown, by ultraviolet absorption spectra, that it is possible to reduce the niacin in coenzyme II by chemical means (*29*). Ordinary heat processing has little effect on the niacin in foodstuffs (*8*).

Niacin is part of the coenzymes I and II and may have significance from a clinical standpoint in the treatment of cancer.

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TABLE 1

Effect of High-Voltage X-Rays on Pure Solutions of U.S.P. Niacin

Concentration..... 100 γ/ml
Volume irradiated..... 3 ml
Voltage..... 3 megavolts

Sample	*Current microamperes*	*Time of Irradiation seconds*	*Dosage*		*Niacin Concentration after Irradiation γ/ml*
			Total roentgens	*Rate r/sec*	
I	230	193	125,000	648	104
G	190	424	250,000	590	104
C	240	545	375,000	688	105
E	240	625	500,000	800	102
F	200	1800	850,000	473	105
L	Control	....	.....	...	104*

* Not irradiated

Apparatus

A Trump generator operating at three million volts (*23*, *26*, *27*), utilizing the pressure-insulated electrostatic principle of Van de Graaff, was used for the production of these radiations. A specially designed X-ray tube was used, which was provided with a window of thin aluminum to close off the tube so that a vacuum could be maintained. Electrons were incident on a water-cooled gold target, 0.25 inch in thickness. The roentgen rays were transmitted in the direction of the electron stream to the sample being irradiated, 3.2 cm below. The X-rays obtained varied in wavelength from 0.004 to 0.06 A.U., with a maximum intensity at 0.008 A.U.

Samples to be irradiated by X-rays were placed in small, cylindrical, stainless steel plates similar to Petri dishes.* These dishes were 0.8 cm high and 2.9 cm in diameter. The covers were 0.5 cm in depth and 3.0 cm in diameter. The dishes were placed in a small adapter, 3.2 cm below the target. The capacity of the dishes was slightly under 3 ml.

For cathode-ray irradiation, the gold target was removed from the path of the electrons. The electrons passed through the thin aluminum window, and cathode rays (beta rays) emerged. The sample was placed in the center of the base of a larger, stainless steel cylinder, 4 inches in diameter, 50 cm below the aluminum plate. The cover of the dish used for cathode-ray exposure was made of thin aluminum and was 0.5 cm high and 3 cm in diameter.

The dosages given are expressed in roentgens, based on ionization of air.

Methods of Assay Used

Niacin was determined photometrically (*16*) in a Cenco Photelometer by a modification of the Mueller and Fox method (*21*) involving the reaction of niacin with cyanogen bromide and ammonium hydroxide. This method was found to be rapid and accurate and to give reproducible results.

Reduced ascorbic acid was determined photometrically, in an Evelyn colorimeter, by the method of Hoch-

* Control tests with glass dishes indicated that the metal containers, which were more convenient to use, played no rôle in the changes observed with niacin during irradiation.

berg, Melnick, and Oser (*17*).

Ultraviolet absorption spectra were determined in a Beckman spectrophotometer. Readings were made at intervals of 5 mμ except at the peaks, in which case the intervals were 1 mμ.

Experimental

Effects of different dosages of high voltage X-rays on niacin in pure solution:

A stock solution of U.S.P. niacin in absolute ethanol containing 100 micrograms of niacin per ml was made and used in this and subsequent experiments. Dilutions were made with distilled water, when necessary.

Niacin concentration 100 micrograms per ml. Samples of the stock solution (3 ml each) were pipetted into the X-ray dishes described above and irradiated for varying periods of time, to obtain total dosages of from 125,000 to 850,000 r (roentgens). After irradiation, the samples were assayed for niacin (Table 1). Ultraviolet absorption spectra were run on an unirradiated control sample and an identical sample irradiated for 850,000 r. Both samples had the same absorption spectra.

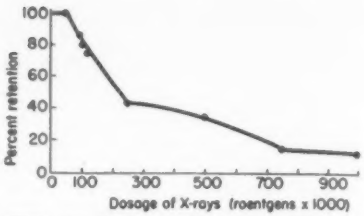

FIG. 1. Effect of different dosages of high-voltage X-rays on retention of niacin in pure solution (50 γ/ml)

Niacin concentration 50 micrograms per ml. Samples (2.5 ml each) of a solution of niacin containing 50 micrograms per ml were pipetted into several exposure dishes and irradiated by X-rays in dosages varying from 50,000 to 1,000,000 r. The irradiated samples and a control sample were then assayed for niacin by the method outlined previously. The irradiation data and the

TABLE 2

Effect of Different Dosages of High-Voltage X-Rays on Pure Solutions of Niacin

Concentration..... 50 γ/ml
Volume irradiated.....2.5 ml
Voltage..... 3 megavolts

Sample	*Current microamperes*	*Time of Irradiation sec*	*Dosage*		*Niacin after Irradiation*	
			Total roentgens	*Rate r/sec*	*Concentration γ/ml*	*Retention %*
6′	60	85	50,000	589	50.0	100.0
15	60	169	100,000	591	43.0	86.0
1	70	142	110,000	770	40.0	80.0
14	70	157	125,000	795	37.5	75.0
17	260	157	250,000	1591	21.5	43.0
28	220	285	500,000	1755	17.0	34.0
30	250	400	750,000	1875	7.3	14.6
6	230	578	1,000,000	1730	6.0	12.0
G	Control	....	.....	....	49.0*	....

* Not irradiated

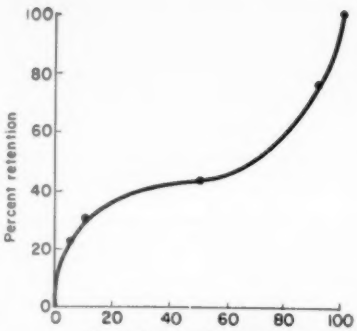

FIG. 2. Effect of dilution on niacin retention after irradiation by high-voltage X-rays (250,000 roentgens)

assay results are presented in Table 2 and depicted graphically in Fig. 1.

Niacin concentration 5 micrograms per ml. In this experiment the concentration of niacin was lowered from 100 to 5 micrograms per ml, and samples were irradiated to the extent of 250,000 r. The results for this dilution are compared in Table 3 and Fig. 2 with those obtained with the higher concentrations of niacin.

Effects of different rates of dosage. To determine whether different rates of X-ray dosage have different destructive effects on niacin when the total dosage is kept constant, 2.5-ml samples of niacin in concentrations of 50 micrograms per ml were irradiated with total dosages of 250,000 r at rates of 295 and 1591 r per sec, and with total dosages of 125,000 r at rates of 177 and 227 r per sec. The data obtained are presented in Table 4.

Effects of different dosages of high-voltage X-rays on niacin in the presence of other compounds:

Forssberg (*10*), Dale (*5*), Tytell and Kersten (*28*), and others have shown that enzymes and organic compounds are "protected" when irradiated by X-rays in the presence of other compounds. The following experiments were conducted to determine whether niacin may be "protected" in a similar manner.

Addition of methionine. Samples (2.5 ml each) of a mixture containing 16 micrograms per ml of methionine and 20 micrograms per ml of niacin and a 2.5-ml control sample of a solu-

TABLE 3

Effect of Dilution on Destruction of Pure Solutions of Niacin by High-Voltage X-Rays

Volume irradiated..... 2.5 ml*
Voltage..... 3 megavolts
Total dosage..... 250,000 roentgens

Sample	*Current microamperes*	*Time of Irradiation sec*	*Rate of Dosage r/sec*	*Niacin Concentration*		*Retention of Niacin After Irradiation %*
				Before Irrad. γ/ml	*After Irrad. γ/ml*	
I	230	193	1294	100	104	100.0
2	230	200	1250	90	64.8	72.0
17	260	157	1594	50	21.5	43.0
1	220	145	1724	10	3.0	30.0
30	230	175	1430	5	1.1	22.0

* Volume of Sample I irradiated was 3 ml

TABLE 4

Effect of Different Rates of Dosage of High-Voltage X-Rays on Niacin in Pure Solutions

Concentration..... 50 γ/ml
Voltage..... 3 megavolts

Sample	Current microamperes	Time of Irradiation sec.	Dosage		Niacin after Irradiation	
			Total roentgens	Rate r/sec	Concentration γ/ml	Retention %
1	50	708	125,000	177	37.5	75.0
24	100	538	125,000	227	37.5	75.0
4	90	849	250,000	295	21.25	42.5
17	260	157	250,000	1591	21.5	43.0
16	Control	...	.....	....	50.0*	....

* Not irradiated

tion containing 20 micrograms per ml of niacin alone were irradiated by 250,000 r. Niacin determinations were made after irradiation. (See Table 5.)

Addition of ascorbic acid. As niacin is a stable molecule chemically and radiobiologically, it was of interest to note what would be the effect of irradiation of a solution of niacin containing ascorbic acid. Therefore, 2.5-ml samples of solutions containing 50 micrograms per ml of niacin and 500 micrograms per ml of U.S.P. ascorbic acid (in 0.5 percent oxalic acid) were irradiated by 125,000 and 250,000 r. Solutions containing 50 micrograms per ml of niacin alone in oxalic acid and 500 micrograms per ml of ascorbic acid alone in oxalic acid were also made, and 2.5-ml samples of these solutions were irradiated by the same dosages. The mixtures were then assayed for niacin and ascorbic acid contents. (See Table 6.)

Ascorbic acid and different quantities of niacin. To determine the minimal amounts of niacin that will "protect" ascorbic acid from X-ray irradiations,

TABLE 5

Effect of High-Voltage X-Rays on Niacin Irradiated in Presence of Methionine

Volume irradiated..... 2.5 ml
Total dosage..... 250,000 roentgens
Voltage..... 3 megavolts

Sample	Concentration of		Current microamperes	Time of Irradiation sec	Rate of Dosage r/sec	Niacin after Irradiation	
	Niacin γ/ml	Methionine γ/ml				Concentration γ/ml	Retention %
24	20	16	200	195	1281	4.25	21.2
2	20	..	230	166	1508	5.2	26.0
Control	20	16	...	...	....	20.0*	100.0*

* Not irradiated

TABLE 6

Effect of High-Voltage X-Rays on Solutions of U.S.P. Niacin, U.S.P. Ascorbic Acid, and on Mixtures of the Two

Voltage..... 3 megavolts
Current..... 170 microamperes

	Concentration of		Time of	Dosage		Concentration after Irradiation		Retention after Irradiation	
Sample	Niacin γ/ml	Ascorbic Acid γ/ml	Irradiation sec	Total roentgens	Rate r/sec	Niacin γ/ml	Ascorbic Acid γ/ml	Niacin %	Ascorbic Acid %
4	50	500	...	Control	...	50.0*	490.0*	....	....
6	50	500	205	125,000	599	22.0	419.0	44.0	83.8
16	50	...	287	125,000	436	43.0	.....	86.0	....
27	..	500	270	125,000	463	...	333.0	....	66.6
15	50	500	542	250,000	462	12.5	392	25.0	78.4
17	50	...	542	250,000	462	20.0	.....	40.0	....
1	..	500	500	250,000	453	....	279	....	55.8

* Not irradiated

2.5-ml samples of mixtures containing 500 micrograms per ml of ascorbic acid and 33.3, 25, or 10 micrograms per ml of niacin, respectively, were irradiated by 125,000 r. (See Table 7.)

Effect of high-voltage cathode rays on niacin in pure solution:

Different times of irradiation. Four small, stainless steel dishes containing 3 ml of a solution of U.S.P. niacin (100 micrograms per ml) were irradiated by cathode rays produced at 3 million volts and 10 microamperes for 15, 30, 45, and 60 sec. Physical ionization measurements show that irradiation for 60 sec at 10 microamperes is equivalent to approximately 3,400,000 r. Niacin determinations were made after

TABLE 7

Effect of High-Voltage X-Rays on Ascorbic Acid Combined with Different Amounts of Niacin

Voltage..... 3 megavolts
Current..... 180 microamperes
Total dosage..... 125,000 roentgens

	Concentration of				Concentration after Irradiation		Retention after Irradiation	
Sample	Niacin γ/ml	Ascorbic Acid γ/ml	Time of Irradiation sec	Rate of Dosage r/sec	Niacin γ/ml	Ascorbic Acid γ/ml	Niacin %	Ascorbic Acid %
4	33.3	500	163	767	4.6	393	13.8	78.6
6	25.0	500	165	758	3.6	402	14.4	80.4
15	10.0	500	167	749	3.0	392	30.0	78.4

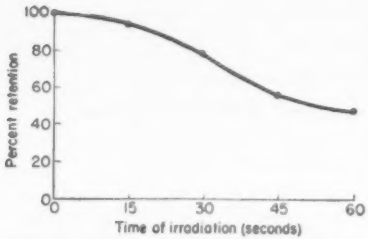

FIG. 3. Effect of high-voltage cathode rays on retention of niacin in solution (100 γ/ml) irradiated at 10 microamperes for different periods

irradiation. (See Table 8 and Fig. 3.)

Dilution effect. To help determine whether the degree of destruction of niacin by cathode rays is more severe in solutions of higher dilution, 3-ml samples of niacin in concentrations of 100, 80, 50, 25, and 10 micrograms per ml were irradiated for 60 sec by cathode rays produced at 3 megavolts and 10 microamperes. Niacin determinations were made after irradiation by the method previously mentioned. (See Table 9 and Fig. 4.)

Effect of high-voltage cathode rays on niacin in the presence of other compounds:

This experiment was designed to determine the relative radiosensitivity of niacin *per se* and in the presence of other compounds. Irradiation for 60 sec with cathode rays at 10 microamperes was carried out on 3-ml samples of solutions, with the results shown in Table 10.

Discussion of Results

Effect of high-voltage X-rays on niacin:

U.S.P. niacin in a concentration of 100 micrograms per ml was not destroyed by X-rays in dosages up to 850,000 r.

When the concentration of niacin in the solution was reduced to 50 micrograms per ml or less, partial destruction

TABLE 8

Effect of High-Voltage Cathode Rays on Solutions of U.S.P. Niacin

Concentration..... 100 γ/ml
Volume irradiated..... 3 ml
Voltage..... 3 megavolts

Sample	*Current microamperes*	*Time of Irradiation seconds*	*Niacin after Irradiation Concentration γ/ml*	*Niacin after Irradiation Retention %*
G	Control	0	100*	100*
E	10	15	94	94
C	10	30	79	79
I	10	45	55	55
F	10	60	46	46

* Not irradiated

was apparent with dosages of 100,000 r or above. The degree of destruction was apparently exponential, as may be seen in Fig. 1.

A similar effect of dilution was noted when solutions of niacin in concentrations ranging from 100 to 5 micrograms per ml were irradiated for 250,000 r.

Niacin is apparently more radiosensitive in dilute solutions than in concentrated solutions. This finding

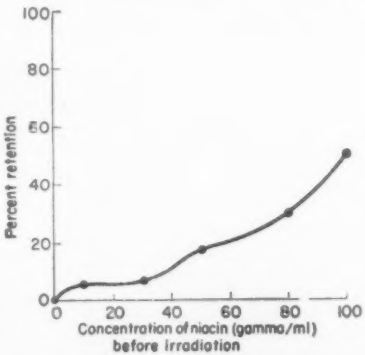

FIG. 4. Effect of cathode rays produced at 10 microamperes for 60 seconds on solutions of niacin of varying concentrations

resembles a radiation phenomenon observed by Dale (*4*) with carboxypeptidase, by Forssberg (*9*) with catalase, and by others.

The results strongly suggest that destruction of niacin by X-rays is the result of indirect action of the radiations on the solvent rather than of direct destruction of the niacin by "hits." The theory is based on the premise that most of the molecules of the solute that react to the radiations have not been excited or ionized directly by the radiations but their reaction follows the ionization of the solvent molecules. In the early literature, this reaction is referred to as "activated water." This theory is in direct contrast to the "hit" or "target" theory or "Treffer Theorie," whereby the molecule or structure in which ionization is produced is designated as the "target" and the production of ionization as a "hit."

If the theory of indirect action, with the production of an intermediary body of finite life, is to be applied to vitamins, there should be a competitive reaction for this intermediary body when a vitamin is irradiated in the presence of another solute. Such has been shown to be the case with catalase by Forssberg (*10*), with carboxypeptidase by Dale (*4*), with acetylcholine by Dale

TABLE 9

Effect of High-Voltage Cathode Rays on Solutions of Niacin of Different Concentrations

Voltage..... 3 megavolts
Current..... 10 microamperes
Irradiation..... 60 seconds

	Concentration of Niacin		
Sample	*Before Irradiation* γ/ml	*After Irradiation* γ/ml	*Retention of Niacin* %
C	100	51.0	51.0
I	80	23.8	29.7
G	50	8.8	17.6
E	25	1.6	6.4
F	10	0.6	6.0

TABLE 10

Effect of High-Voltage Cathode Rays on Niacin Alone and on Niacin in the Presence of Other Compounds

Voltage..... 3 megavolts
Current..... 10 microamperes
Irradiation..... 60 seconds*

	Concentration before Irradiation		*Niacin after Irradiation*	
Sample	*Niacin* γ/ml	*Other compounds*† γ/ml	*Concentration* γ/ml	*Retention* %
G	50	50 G	8.75	17.5
E	42.8	12.9 M	8.75	20.4
F	66.7	33.3 G	13.75	20.9
G′	50	50 C	3.75	7.5
I′	50	50 C′	5.50	11.0
Control 1	50	.....	8.75‡	17.5‡
Control 2	40	.....	14.0‡	10.0‡
Control 3	66.7	.....	14.0‡	21.0‡

* Sample E was irradiated for 62 seconds
† G = glycine; M = methionine; C = cystine; C′ = cysteine · HCl
‡ Not irradiated

(*5*), and with urease by Tytell and Kersten (*28*).

When niacin was irradiated in the presence of methionine, the data show that methionine competed with niacin for the ionized particles, but the niacin was slightly more reactive with the activated water and "protected" the methionine. The difference in niacin destruction when niacin alone was irradiated and when niacin combined with methionine was irradiated was not great enough, however, to justify sweeping conclusions.

When niacin in combination with ascorbic acid was irradiated by 125,000 and 250,000 r, the results were unexpected. Although ascorbic acid is a more labile molecule than niacin and was more labile to X-rays when irradiated alone, when the two combined were irradiated by X-rays, the niacin was destroyed more readily than the ascorbic acid. Hence ascorbic acid appears to be protected by niacin. One may assume that niacin competes with ascorbic acid for the activated water particles in this instance. Niacin in concentrations of as little as 10 micrograms per ml protected 500 micrograms per ml of ascorbic acid from X-rays.

Reference to the previous tables indicates that the rate of dosage of X-rays was not the same in all instances, which presents the possibility that the results obtained might be ascribed to this fact. Accordingly an experiment was conducted in which solutions of niacin of a given concentration were irradiated by X-rays for total dosages of 250,000 and 125,000 r, respectively, each total dosage being delivered at two different rates. This was accomplished by changing the current. For the total dosage of 125,000 r the second of the two rates of dosage was increased 30% over the first rate, and for the total dosage of 250,000 r, 440%. The data show that at a given total dosage the net results in niacin destruction were the same, irrespective of the difference in the rates of dosage, even when this difference was as much as 440%.

Effect of high-voltage cathode rays on niacin:

Cathode rays produced at 10 microamperes for as short a period as 15 seconds were destructive to niacin in pure solution, even at a concentration of 100 micrograms per ml. Hence cathode rays may accomplish in a few seconds what it takes X-rays minutes to do.

In no case of cathode or X-ray irradiation in this investigation was any noticeable heat produced in the samples. This is to be expected, for in an X-ray beam of 1 sq cm there is produced but 10^{-3} to 10^{-5} calorie per sec (*15*).

The amount of destruction of niacin by cathode rays increased with dilution, as in the case of X-rays. Therefore, it would seem that the action of cathode rays on niacin is indirect.

Curves for the ultraviolet absorption spectra of solutions of niacin (100 micrograms per ml), irradiated for 45 and 60 seconds by high-voltage cathode rays at 10 microamperes, are presented in Fig. 5. Irradiation, which resulted in a loss of niacin as represented by the cyanogen-ammonia reaction, also resulted in a slight shift in the absorption maximum as compared with the absorption maximum of nonirradiated niacin, namely, from 262 to 263 mμ. The minimum changed from 238–239 to 241 mμ. When niacin was irradiated, the optical density increased from 270 mμ on. These data confirm the assay results and, furthermore, may offer some possible means of detecting what happens to the niacin molecule upon irradiation, for microchemical methods are not feasible with such small quantities. Further work on this is in progress.

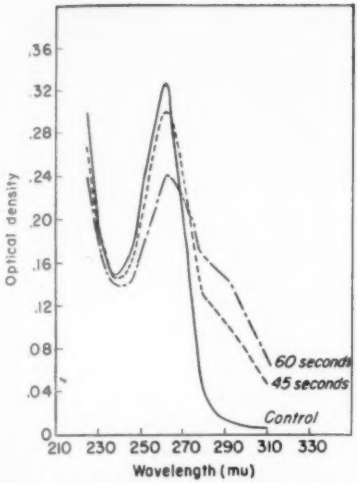

FIG. 5. Effect of high-voltage cathode rays on ultraviolet absorption spectrum of niacin in solution (100 γ/ml) irradiated for 45 and 60 seconds at 10 microamperes

Methionine protected niacin from cathode rays. There was a 20% retention of niacin when it was irradiated in the presence of methionine and only a 10% retention when niacin was irradiated alone. Glycine offered no protection to niacin from cathode rays. There was a greater destruction of niacin when it was irradiated in the presence of cysteine or cystine than when irradiated alone by cathode rays.

Summary and Conclusions

1. Pure solutions of niacin were irradiated by X-rays and by cathode rays produced at 3 megavolts, and the degree of retention of niacin after irradiation was measured.

2. Niacin in a concentration of 100 micrograms per ml was not destroyed by X-rays in total dosages between 125,000 and 850,000 r.

3. When the concentration of niacin was reduced to 50 micrograms per ml, partial destruction of the vitamin occurred at X-ray dosages between 50,000 and 100,000 r.

4. The percentage destruction of niacin of this concentration was increasingly greater with increasing dosages of X-rays between 50,000 and 250,000 r, but at greater dosages the destruction decreased, percentagewise, until it reached a constant level at dosages between 750,000 and 1,000,000 r.

5. When the total X-ray dosage was 250,000 r, the percentage destruction of niacin was greater for greater dilution of vitamin, but the increased destruction was not linear with the dilution.

6. The effect produced on niacin by a given total dosage of X-rays was independent of the rate of dosage up to at least five times the initial rate.

7. When niacin was irradiated by X-rays in the presence of methionine, niacin loss was slightly greater than that when niacin was irradiated alone.

8. Niacin in the presence of ascorbic acid was destroyed by hard X-rays more readily than niacin irradiated alone, although the ascorbic acid alone was more radiosensitive than the niacin alone. In the presence of niacin, ascorbic acid was less radiosensitive to hard X-rays.

9. Ascorbic acid (500 micrograms per ml) was protected from hard X-rays by as little as 10 micrograms per ml of niacin.

10. High-voltage cathode rays had a destructive effect on niacin in a concentration of 100 micrograms per ml in as short a period of irradiation as 15 seconds at 10 microamperes.

11. Increased time of exposure of niacin in this concentration to cathode-ray irradiation at 10 microamperes resulted in increased destruction of the niacin. The rate of destruction was not linear, however, with the time of irradiation.

12. Dilution of the niacin solutions irradiated by high-voltage cathode rays had the same effect on the retention of

niacin as was noted in the case of X-ray irradiation of dilute solutions. The relationship between the dilution and the retention of niacin was not linear, but at low concentrations was asymptotic.

13. There was a greater retention of niacin when it was irradiated by cathode rays in the presence of methionine than when it was irradiated alone. The reverse held true when niacin was irradiated with cysteine and cystine.

14. The action of both hard X-rays and cathode rays on niacin appeared to be indirect.

15. Irradiation of niacin by cathode rays resulted in a change in the ultraviolet absorption spectrum.

* * *

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Problems of Radioactive Waste Disposal*

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OUR PRESENT KNOWLEDGE of radioactive waste disposal is quite small compared to what remains to be learned about the subject. In fact, in many cases, it is thought of as the radioactive waste disposal *problem*.

In this paper, the objectives are:

1. To indicate the nature of the problem; and
2. To mention merely the general principles by which we may expect the problem eventually to be solved.

The problems of radioactive waste disposal are not entirely different from some of the problems of industrial hygiene. Radioactive waste products are, first of all, chemicals. As such, they have approximately the same chemical behavior as have the stable isotopes of the same chemical elements. Secondly, radioactive toxicity creates problems similar to those of chemical toxicity. In work with radioactive materials, the ideal objective is to retain absolute control of all radioactive wastes. However, since this is generally impossible, the practical goal is that of preventing damage to human tissues. (In some cases, the probability of economic waste may be a dominant consideration.) In this comparison, perhaps the most significant differences between chemical toxicity and radioactive toxicity are the nature of the results and the greater lengths of time required for the results of radioactive toxicity to become evident.

Many of the elements of radioactive toxicity are called to mind by the statement that, as health hazards, radioactive materials have a well-known prototype in radium, which, more than twenty years ago, was identified as the responsible agent in the occupational poisoning and subsequent deaths of

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TABLE 1

Example Illustrating Differences in Specific Radiotoxicity Due to Differences in Physical and Chemical Properties

Property	*Ra^{226} and its radioactive products*	*C^{14} (CO_2)*
Types of radiation	Alpha, beta, gamma	Beta
Average energy, per disintegration of Ra^{226} or C^{14}	14 Mev*	0.05 Mev
Radiation half-life	1,600 years	5,000 years
Organ most affected	Bone	Soft tissues† of whole body
Fraction of inhaled isotope fixed in organ	0.25	0.25
Elimination rate	0.01% per day (T_e = 20 years)‡	7% per day (T_e = 10 days)‡
Tolerance concentration in air (2,000 hours/year)	1×10^{-10} gm/cc	1×10^{-7} gm/cc

* It is assumed here that approximately half of the radon formed in the body as the result of disintegration of the radium escapes. The total energy of disintegration, per radium atom disintegrating, of radium and its short-lived products is: alpha, 25 Mev; beta, 1.3 Mev; gamma, 2 Mev.

† There is some evidence that under certain circumstances, important quantities of C^{14} may be deposited in bone with a very low elimination rate.

‡ T_e is the time required for any quantity deposited in the tissues to be reduced to half through elimination. Exponential elimination is assumed. No claim is made for the accuracy of the numerical values given here.

many workers in the radium industry. The physical properties of radium which account for its toxicity are shared, in some degree, by all radioactive materials. Atoms of radioactive isotopes disintegrate with the release of amounts of energy which, compared with those resulting from chemical processes involving the same numbers of atoms, are extremely large.

When such radioactive atoms are within body tissues, this released energy is spent in breaking down the tissues. In general, all of the energy of the emitted particles and much of the energy of the emitted gamma radiation is absorbed in the surrounding tissues. As a result of these types of radiation, if the quantities of radioactive materials in an organ of the body were sufficiently large, very rapid failure of that organ would result. In more chronic cases, we risk various types of malignancy or, at best, marked reduction in vigor.

Although radioactive toxicity depends upon the general properties common to all radioactive atoms, there are significant differences between the various isotopes. Physical properties which differ include the types of radiation involved in disintegration, the energies associated with the radiated particles and quanta, and the half-lives or decay constants of the radioactive materials. Chemical properties of interest are those which account for the fractions of ingested or inhaled materials reaching the various organs of the body and for the rates of elimination of these materials from the organs of the body.

It is, perhaps, worthwhile to compare, as an example, two radioisotopes of very different physical and chemical properties. Table 1 gives such a comparison for radium and a radioactive isotope of carbon, C^{14}. The effects indicated for radium include those of a large fraction of its short-lived radiation products

which are assumed to be retained in the body until after their decay.

An inspection of the table shows the average energy per disintegration from radium and its products to be extremely large compared to the average energy per disintegration from C^{14}. Moreover, alpha particles are believed to produce some ten times as much tissue damage as beta particles of the same energy. The next most significant difference between these two radioisotopes is in their elimination rates. Whereas it is estimated that only 10 days is required for any amount of C^{14}, fixed in the soft tissues of the body, to be reduced to half by elimination, some 20 years, or more, may be required for a similar reduction in radium. As long as the radioisotope is in the tissue, the radiation continues at a rate proportional to the remaining amount. Principally as a result of the differences pointed out in this discussion, the permissible amount of C^{14} in air is estimated to be of the order of 100,000 times that of radium.

With this introduction to the nature of the hazards which result from the presence of radioactive wastes in materials taken into the body, we will consider next other elements of the problem of disposal of radioactive wastes. Interest in this problem has been greatly stimulated, during the past two or three years, by activities associated with the production and use of fissionable materials. For the present discussion, our interests in fissionable and radioactive materials may be considered as falling into three groups:

1. Operations incidental to the production of fissionable materials and other radioisotopes. These include the processing of uranium and other strategic ores for the production of fissionable materials.
2. The use of radioisotopes for various experimental, and other, purposes.
3. The use of fissionable materials or fission products in warfare.

Under peacetime conditions, by far the greatest amounts of radioactivity involved in nuclear operations are associated with the operation of chain-reacting piles and associated chemical processes. Table 2 gives, as an example, the activities of a few of the fission products to be found in the metal of a small chain-reacting pile after 100 days of continuous operation.

Relationships between the activity, half-life, and mass of three radioisotopes are illustrated in Table 3.

The number of atoms of a particular isotope required to make a curie is proportional to the half-life of the iso-

TABLE 2

Activities of Certain Long-Life Fission Products in Pile After 100 Days

Isotope	*Half-life**	*Percent fission yield**	*Activity (curies)*
Sr^{89}	53 d	4.6	28,600
Sr^{90}	25 y	Assumed 5	330
Y^{91}	57 d	5.9	35,400
Ru^{106}	1 y	0.5	740
Te^{132}	77 h	3.6	30,700
I^{131}	8 d	2.8	23,900
Cs^{135}	20,000 y	Assumed 5	50

* These values are from *J. Am. Chem. Soc.* **68**, 2411 (1946).

TABLE 3

Illustration of Relationships Between Activity, Half-Life, and Mass

Isotope	*Half-life*	*Gm/curie*	*Curies/gm*
Ra^{226}	1600 y	1 (by definition)	1
Cs^{135}	20,000 y	7.5	0.13
I^{131}	8 d	8×10^{-6}	120,000

TABLE 4

Decay of Certain Long-lived Fission Products with Time

Isotope	*Half-life*	*Curies* (T = 0)	*Curies* (T = 100 *days*)	*Curies* (T = 1 *year*)	*Curies* (T = 10 *years*)
Sr^{89}	53 d	28,600	7,700	240	2×10^{-21}
Sr^{90}	25 y	330	327	321	250
Y^{91}	57 d	35,400	10,500	420	—
Ru^{106}	1 y	740	610	370	0.7
Te^{133}	77 h	30,700	0.1	—	—
I^{131}	8 d	23,900	5	—	—
Cs^{135}	20,000 y	50	50	50	50

tope; and the number of grams of the isotope required to make a curie is proportional both to the half-life and to the atomic weight. Appreciation of these relationships is essential to an understanding of many waste disposal problems, since frequently the masses involved in tremendous amounts of radioactivity are so small that only the principles of trace chemistry apply.

The activities of the radioisotopes listed in Table 2 total 120,000 curies. Although this is but a small fraction of the total activity due to the 160 fission products in the pile, it is perhaps one hundred times that of all of the radium produced before 1942. Such a comparison really has little meaning, however, for, not only is the average hazard per curie of these radioisotopes less than that for radium, but nearly all of these isotopes are relatively short-lived and will have almost entirely decayed to nonradioactive isotopes within a few months or years. This is illustrated by Table 4. Many fission products have half-lives measurable in seconds, minutes, or hours, and decay far more rapidly than those listed in the table.

We are now in a position to consider how some of the problems in radioactive waste disposal arise. Whether one operates a chain-reacting pile for the production of plutonium, for the production of radioisotopes, for the production of power, or for experimental purposes, eventually there comes a time when it is desirable to separate, chemically, various radioisotopes from the mixture of partially depleted uranium and its products. Such separations ordinarily involve the use of comparatively large amounts of liquid carrying considerable quantities of radioisotopes in solution. Some operations involve the production of considerable quantities of radioactive gases or of air-borne radioactive materials. As is true in any chemical plant, it is necessary to determine that liquids released from such an operation cannot be injurious in any way to human life, and will not represent any great economic liability. Specifically, the radioisotopes released into the drainage system or into the atmosphere must be incapable of sufficient concentration in air, water, food, or drugs to be harmful to man, or to plant and animal life.

The question of neutralization of radioactive materials arises many times. However, such materials cannot be neutralized. The basis for this statement is twofold: (1) For our purpose, there is no practical method of changing the radioactive properties of a large fraction of any of the radioisotopes in the waste; and (2) medical research has not discovered any treatment which is known to neutralize the tissue damage

TABLE 5

Tolerance Concentrations of Various Radioisotopes in Drinking Water

	Absorption in critical tissue (%)	*Critical tissue*	T_e *(days)*	T_r *(days)*	T *(days)*	E *(Mev)*	*Ingestion tolerance (μc/liter)*	*Ingestion tolerance (μgm/liter)*
Sr^{89}	7.5	Bone	200	53	42	0.6	4.0	1.4×10^{-4}
Sr^{90}, Y^{90}	7.5	Bone	200	9100	196	0.9	0.58	3.5×10^{-5}
Y^{90}	0.14	Bone	500	2.5	2.5	0.7	3100	5×10^{-3}
Zr^{95}, Cb^{95}	0.005	Bone	80	65	36	2.6	1600	7×10^{-2}
Cb^{95}	0.2	Bone	50	35	21	0.8	240	6×10^{-3}
Ru^{103}	0.00015	Kidney	20	42	14	0.8	11,000	5.5×10^{-1}
Te^{127}	1.5	Kidney	15	90	13	0.23	4.0	3.4×10^{-4}
Te^{129}	1.5	Kidney	15	3.2	2.6	1.7	2.7	8×10^{-6}
I^{131}	20	Thyroid	30	8.0	6.3	0.57	0.06	5×10^{-7}
I^{133}	20	Thyroid	30	0.1	0.1	1	0.14	1.4×10^{-7}
Cs^{137}	30	Muscle	10	12,000	10	0.97	5.4	6.5×10^{-2}
Ba^{140}, La^{140}	6	Bone	200	13	12	2.76	3.7	5×10^{-5}
Ce^{141}	0.18	Bone	50	38	18	0.43	560	1.7×10^{-2}
Ce^{144}, Pr^{144}	0.18	Bone	50	275	42	1.20	80	2.4×10^{-2}
Pr^{143}	0.002	Bone	40	13.8	10	0.3	130,000	2

Explanation of terms: T_e is time required for reduction to half by elimination; T_r is radioactive half-life, and T is resultant time for reduction to half in tissue. E is the average energy released per disintegration. Tolerances are expressed both in microcuries per liter of water and micrograms per liter of water. Tolerance estimates given are tentative because of uncertainty in biological data on which they are based.

produced by such isotopes after they are fixed in body tissues. We cannot overlook, of course, the possibility that such medical advances will be made in the future. However, for the present, we must consider means by which harmful concentrations of radioisotopes can be kept from animal and plant life.

Although many details of the problem of waste disposal remain to be worked out, some of the possibilities are readily evident:

1. Let us consider first the possibility of rendering radioactive wastes harmless by dilution. To the extent that we can determine the maximum quantities of radioactive isotopes which may be taken into the human body under various conditions, without injury, we might expect to be able to use this knowledge as a guide to the concentrations we could tolerate. Table 5 gives estimates of concentrations of various radioisotopes which might be tolerated singly in drinking water for many years. Some of these estimates are only tentative, since they depend upon questionable biological data. The treatment of a case where two or more isotopes are present in the water may be inferred from the statement that the radioactive effects of such isotopes, uniformly distributed through the same tissue, are presumed to be additive.

The third column of Table 5 gives the critical tissues for the particular radioisotope, that is, the tissue which will receive the greatest dosage from that isotope. This is one of the factors which determines the amount of the radioisotope which can be taken safely into the body, since the amount which can be tolerated is proportional to the weight of tissue in which it is distributed.

The fourth, fifth and sixth columns give quantitative factors upon which tolerance values depend. The permissible concentration of the radioisotope is inversely proportional to the magnitude of each of these factors.

The effects of these various factors are readily apparent from an examination of the table. For example, comparing the entries for Sr^{90} with those for Sr^{89}, we see that Sr^{90} gives a tolerance concentration with only 1/8 as many microcuries per liter as are required for Sr^{89}. This is principally due to the longer half-life of Sr^{90}, although the greater energy of disintegration of Sr^{90} (including the energy of disintegration of its short-lived product, Y^{90}) is a contributing factor. Note, incidentally, that if one expresses tolerance concentrations in micrograms per liter instead of microcuries per liter, the number required for Sr^{90} is about 20 times that required for Sr^{89}. Since Sr^{90} has a half-life of 25 years and the half-life of Sr^{89} is only 53 days, the number of grams of Sr^{90} required to produce a curie of activity is about 175 times the number required for Sr^{89}.

Considering further examples from the table: The large activity of Ru^{103} estimated to be required to give a tolerance concentration is based on the very low value reported for absorption of ruthenium from the digestive tract, whereas the very low tolerance value of I^{131} is due to a combination of high absorption and small weight of thyroid tissue in which the iodine concentrates.

In considering the data of Table 5, it is appropriate to re-emphasize the very small concentrations of many of the radioisotopes required to produce tolerance dosage. Further a distinction between estimated tolerance concentrations and operating practices should be made. The values given in the table make no allowance of any kind for uncertainties in our knowledge of the factors upon which the estimates are based. To accept these values as a basis for operating practice, without the introduction of liberal safety factors, would be very unwise.

Suppose now, that we are draining into a sewer, stream, or bay, radioactive wastes sufficiently diluted that we could safely use the water, if otherwise purified, for drinking. Can we assume that these radioactive wastes will remain so diluted? Nature frequently shows remarkable abilities to concentrate various chemical elements especially when they are present only in trace amounts as is the case with many of our radioisotopes. For example, a small bit of a certain species of algae growing in a pool containing such wastes may concentrate them to the extent that the radiation from the algae may be detected at a distance of many feet. Clays and other minerals strongly absorb certain radioisotopes; and, at certain pH values, some resins quantitatively remove them from solution. Such phenomena lead many well-informed persons to question whether radioactive wastes can be made safe for disposal in a drainage system, or in the ocean, merely by aqueous dilution.

2. A second type of dilution, however, can perhaps be used in safety wherever it is practical. If the radioisotope is diluted by and uniformly mixed with one or more stable isotopes of the same chemical element, to the point where plant or animal life cannot take it up in sufficient quantities to present a radioactive hazard, there is no reason to expect any considerable subsequent separation in nature.

3. A review of Table 4 suggests a very useful principle in the handling of radioactive wastes. If we can find some safe place to store the waste for a considerable time, a large portion of the radioactive material can decay into stable isotopes without any chance of producing damage. Obviously, applications of this principle are limited by the half-lives of the radioisotopes involved and by the availability of suitable storage space. When one starts to apply this method to such long-lived isotopes as Cs^{135} and Pu^{239},

with half-lives of 25,000 years and 24,000 years, respectively, one must consider what sort of container may be expected to endure for many thousands of years.

4. We have noted that the principle of control of radioactive wastes by decay is limited by storage space. For plants working with radioactive materials on a large scale, much consideration is being given to concentration of these materials by removal from the process fluids. Although detailed processes for such removal have not been developed, there appears to be no reason to suppose that such development will be too difficult. Methods of concentration from waste water, frequently discussed, involve such principles as those of ion exchange, adsorption, precipitation, sedimentation, solvent extraction, and evaporation.

5. Once the radioactive wastes are concentrated to convenient volumes, current thinking appears to be divided between two courses of action. Some groups talk of placing them in storage for indefinite periods of time, until national or international policies of waste disposal can be established. Other groups plan to dispose of them, in suitable containers, in one of the oceans. Perhaps the best method so far proposed for such disposal is that of using the concentrated solution as the liquid for making solid concrete blocks which can be dumped into the ocean with assurance that the major portion of the radioactive material will sit in one place for a long time. However, for such long-lived elements as plutonium, some persons feel that even this method leaves something to be desired. There may also be practical difficulties, since some wastes contain sufficient quantities of certain chemicals to prevent setting of the concrete.

An important feature of any considerable waste disposal program is the monitoring of discharged air and water to assure that their radioactive contents do not become excessive. This subject is too extensive to include in the present discussion.

We have devoted a great deal of time to the discussion of the disposal of liquid wastes from radioactive chemical plants, partly because they constitute one of the principal problems of waste disposal in time of peace, and partly because the problems considered are quite similar to those met in other activities involving the use of radioisotopes. We shall pass over the problem of disposal of small amounts of radioisotopes used in various types of experimental work by saying that a minimum precaution to observe, in letting them out of the custody of a responsible person, is that of determining that they are in too dilute concentrations to be harmful to life. In the case of short-lived radioisotopes, the most convenient procedure is to allow them to decay to negligible activities.

The problem of radioactive contamination in the event of a war involving the use of atomic bombs and, perhaps, other types of radioactive weapons, is readily apparent. However, the problem of control of such contamination lies outside the field of the present discussion.

Theory and Operation of Geiger-Müller Counters—II

COUNTERS FOR SPECIFIC PURPOSES

Details of construction are presented for various types of counters, including those for gamma, beta and X-rays, for photons, for liquid and gaseous sources, and for various special purposes. Advantages and disadvantages are cited

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THE PROPERTIES of the types of ionizing radiation which may be detected with Geiger-Müller counters are so different that there is no such thing as a universally efficient tube. So far as the characteristics of the radiation overlap one another, the same counter construction may be used to detect them, but it is more efficient to design an optimum construction for each detection problem.

In many cases, the extra work involved in designing and constructing an optimum counter is not worth the increase in efficiency obtainable and, therefore, it is quite common to consider only two general types of detectors, gamma-ray and beta-ray counters.

The purpose of this paper is to discuss briefly a number of counters which have been designed and tested by workers in the field of radioactivity detection. No attempt is made to cover all possible modifications. Each one serves as an example of a type useful in certain specific circumstances and could serve as a suggestion for optimum efficiency in a particular detection problem.

GAMMA-RAY COUNTERS

For Various Energies

The easiest type of Geiger-Müller counter to construct is the general purpose gamma-ray tube. Since gamma rays are very penetrating, the details of the envelope can be designed for ease in construction. For electromagnetic energies higher than about 50 kilovolts, the absorption in any practical counter-filling gas is negligible, and hence, the gas filling can be chosen with consideration for the discharge mechanism alone. Although the secondary electrons produced by the gamma rays from the cathode walls are the ionizing particles which initiate the count, unless one is interested in particularly high efficiency even the material of the cathode walls is no great factor. A typical example of a simple gamma-ray counter is shown in Fig. 1.

If the highest possible efficiency is desired for the detection of gamma radiation, the material of the cathode is all-important. Secondary electrons are produced by gamma rays in three ways: through photoelectric absorption, by Compton scattering, and by pair production. The photoelectric absorption coefficient is approximately proportional to the cube of the atomic number and decreases rapidly with increasing gamma-ray energy. The Compton scattering coefficient is essentially independent of atomic number and decreases slowly with increasing gamma-ray energy. Pair production takes place only for gamma-ray energies above 1.02 Mev energy. The ab-

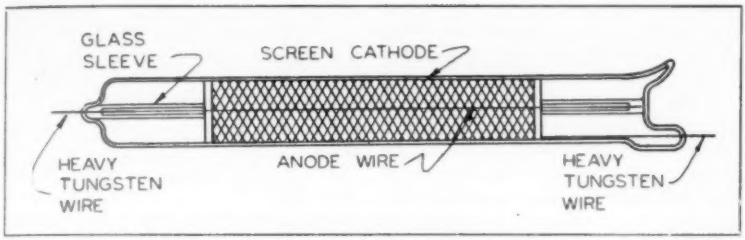

FIG. 1. A screen-cathode gamma-ray counter

sorption coefficient for it is approximately proportional to the atomic number, and increases slowly with increasing energy.

Thus, the energy dependence of these three types of gamma-ray absorption is very different, and their relative importance depends strongly on the atomic number of the counter cathode. In general, at low energies and again at very high energies, a heavy-element cathode is considerably more efficient than one made of a light element. It is approximately true that the efficiency of a counter made of copper or other light element, is proportional to the gamma-ray energy. For a bismuth counter, the efficiency is approximately constant below one Mev and then rises about like that of a copper cathode. Behavior of the efficiency of these two types of counters is illustrated in Figs. 2 and 3 (*1*, *2*).

Although the construction of cathodes of copper and similar materials presents no problem, the manufacture of bismuth cathodes requires a special technique which has been described by Good, Kip, and Brown (*3*). Bismuth is very brittle, has a low melting point, is not available in sheets, and cannot be successfully soldered or spot welded. Satisfactory cathodes are made by plating the bismuth on copper screen.

Since the gamma ray is detected by the counter only through the secondary electrons produced at the cathode walls, it is only the thin layer of the counter wall facing the sensitive volume which is effective in producing counts. Thus the efficiency of the gamma-ray counter is approximately proportional to the area of the solid material in the counter.

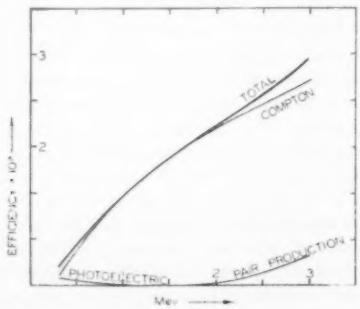

FIG. 2. Efficiency of a copper-wall, gamma-ray counter

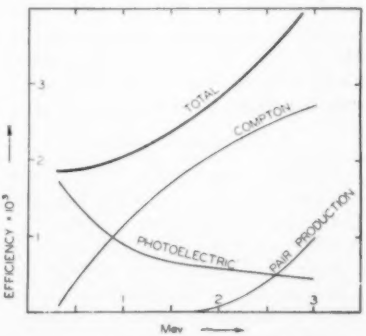

FIG. 3. Efficiency of a bismuth-wall gamma-ray counter

Evans and Mugele (4) first pointed out the increase in sensitivity resulting from increasing the effective area by constructing the cathode of wire screen. Another advantage of the screen is that the field of the counter can extend through the screen and pull into the sensitive volume electrons formed on the outside. The area of metal intercepting the gamma-ray beam will increase as the screen becomes finer in mesh. The efficiency will, therefore, increase with the fineness of the screen until the holes are so small that the field no longer extends beyond the physical boundary of the cathode. The optimum mesh is about 80 to 100 wires to the inch.

Other methods, e.g., using corrugated metal cathodes, introducing fins and partitions into the sensitive volume or building banks of cylindrical counters within the same envelope, are used to increase the effective area. Of these possibilities, the introduction of fins or partitions is the most efficient. A sketch of such a construction is shown in Fig. 4. The partitions can be copper disks in which a large central hole is punched to allow passage of the anode wire. This type of counter construction is satisfactory only with polyatomic-gas-filled counters, since it is necessary to restrict the discharge mechanism to the close vicinity of the axial electrode so that the distortion of the cathode field due to its peculiar shape will not destroy the satisfactory counting characteristics.

Directional Counters

Problems frequently arise where counters are needed to detect the position of radiating sources. There are a number of ways that this can be accomplished, and, for a specific application, the usual balance must be achieved between efficiency, resolution and instrumental complexity.

An ordinary gamma-ray counter will

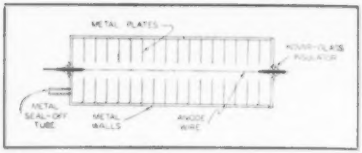

FIG. 4. A high-efficiency gamma-ray counter

show some directional properties because of the fact that a gamma-ray beam approaching the counter from different directions will subtend different amounts of cathode area. In the counter illustrated in Fig. 1, gamma rays coming perpendicular to the axis subtend a much larger cathode surface than an axial ray and hence have a greater efficiency. The reverse is true in the counter illustrated in Fig. 4, where the radial fins increase the probability of production of a secondary electron for a gamma ray entering parallel to the axis, compared to a ray entering perpendicular to the axis.

Some directional properties are also observable because of the interaction mechanism between the gamma ray and the cathode material. For low-energy gamma rays, Figs. 2 and 3 show that the gamma-ray interaction is mainly by the photoelectric effect. Photoelectrons are emitted in a preferred direction at right angles to the gamma-ray beam. Therefore, for low-energy gamma rays, particularly where the photoelectric efficiency is increased by use of heavy-element cylindrical cathodes, greater sensitivity is realized for axial gamma rays than for those entering the counter normal to the axis. The obvious reason is that the more nearly parallel to the cathode surface the gamma ray enters, the more likely the secondary electron will be to enter the sensitive volume of the counter. Although these directional properties are important enough to be considered in reducing background, the resolution is so poor that these mechanisms by them-

selves are not sufficient to produce satisfactory directional characteristics.

The attenuation of low-energy gamma radiation in lead is great enough so that by adding this phenomenon to the angular dependence of the photoelectric effect, one can obtain a reasonably good high-sensitivity, low-resolution directional system. To take a specific example, a platinum cathode gamma-ray counter in a cylindrical lead shield one quarter of an inch thick has a resolution for 0.5 Mev gamma rays such that the counting rate drops to one half when the source subtends an angle of ten degrees with the axis of the counter. The resolution of this system can be greatly improved by using two shielded counters, side by side, separated by a lead shield. The pulses from the two counters are amplified by standard circuits and their outputs subtracted so that if the counting rates are the same from both counters the output reads zero, and if one counter is operating faster, the output can show a large variation from zero. The actual circuits for achieving this will be discussed in the third section of this review series.

The result of this arrangement for a case similar to that taken above, that is, two platinum cathode counters shielded by one quarter of an inch of lead outside and between the two counters, detecting 0.5 Mev gamma rays, gives a resolution such that the differential counting rate rises to one half its maximum value when the source subtends an angle of one degree with the axis of the counter system. This method of achieving directional characteristics is good only for a single point source. If more than a single source is present, the zero counting-rate position will be directed toward an intensity average point such that both counters are receiving the same radiation intensity.

In our discussion of the gas discharge mechanisms in polyatomic gas filled counters,* it was pointed out that the entire discharge, after the initial avalanche, took place in the immediate vicinity of the center wire. One can, therefore, effectively make a series of separate counters all within the same envelope by interrupting the electric field by some method such as sealing glass beads to the center wire, so that the discharge will come up to the glass bead and stop.

As an example, let us choose a gamma-ray counter with a cathode six inches long. On the anode wire, let us seal two glass beads, three millimeters in diameter, spaced so that there are three two-inch sections of center wire along the axis of the cathode. When an electron starts from the cathode toward the anode, it initiates a cumulative ionization avalanche which, when it reaches the vicinity of the anode wire, forms a space-charge-limited discharge which propagates along the wire in either direction, as discussed in the first section of this review series. The diameter of the glass beads on the center wire is greater than the diameter of the propagated discharge, and hence, when this reaches a bead, it is stopped.

This beaded center-wire counter will have directional characteristics, since electrons entering the sensitive volume radially cannot cause a discharge in more than a single section between beads. On the other hand, if an electron travels parallel to the axis of the counter, its secondary electrons will cause avalanches radially, in the field direction, and, provided that the mean free path of the primary electron is at least as great as the length of the sensitive volume, it will produce breakdown in all sections of the counter. Since the size of the pulse from a counter discharge is proportional to the length of the axial electrode, a beaded center-wire counter can easily distinguish

* Nucleonics **2**, No. 6, 14 (1948).

between radial and axial electrons. In this example, electrons coming in the radial direction will cause pulses only one third the size of the pulses coming from electrons traveling parallel to the axis, and this can easily be discriminated by a suitable electronic circuit.

The complication of this method of achieving high-resolution, directional, gamma-ray counters is that of producing secondary electrons in the desired direction. A secondary electron radiator placed at the end of the counter can serve as a source of electrons in the direction parallel to the axis, but the angular distribution of these secondary electrons about the most probable direction limits the resolution of this type of directional gamma-ray counter.

Electronic circuits can be arranged to detect coincident pulses coming from two different counters. Thus, one can set two counters beside each other and design a circuit to detect pulses which come in coincidence and reject all others which do not arrive within the coincidence resolving time of the circuit. Such circuits will be discussed in detail later. By using such an arrangement, one can set up two parallel counters, so that the secondary electron formed at the cathode of one counter can enter the sensitive volume of the second counter and the circuit detect only coincident counts. Since the direction of the secondary electron is given by the primary gamma ray, and the angular resolution for detection is given by the solid angle subtended at the first counter and by the sensitive volume of the second, any degree of resolution can be achieved by adjusting the distance between counters provided the mean free path of the secondary is long enough to allow its entrance into the second counter. The disadvantage of this method is its low efficiency and the instrumental complexity of the two counters and associated electronic circuits.

BETA-RAY COUNTERS

For Various Energies

The major problem in detecting beta-rays is for the particles to penetrate from outside the counter into the sensitive volume. There are two forms of beta-ray counters in general use, one in which the cathode walls are made thin enough to allow the particles to penetrate, the other in which a window is provided for this purpose.

Beta-ray counters which are built to allow the passage of the particles through the envelope walls of the counter are constructed like gamma-ray counters. The glass envelope walls are blown very thin and the cathode, in general, consists of a silver surface deposited on the inside of the envelope. Due to the fact that there is very little material in the counter to act as an efficient secondary electron emitter, the gamma-ray efficiency of such a counter is low and the counter can be made long to insure good plateau characteristics without increasing unduly the gamma-ray background count.

Where samples can be wrapped around the counter, such as sources on filter paper, this type of counter is capable of making use of large source area. It has two principle disadvantages. It does not have maximum efficiency for detecting flat sources, such as deposits on flat plates, since the solid angle of counting a flat source with a cylindrical sensitive volume is not maximum. The more serious disadvantage, however, is the fragility caused by the very thin glass section in the middle of the counter which is necessary to allow the beta rays to enter the counter. For relatively energetic particles, the glass does not have to be too thin and the counter can be very useful. This type cannot be used for the low-energy beta rays since the envelope cannot be made thin enough to allow the entrance of these

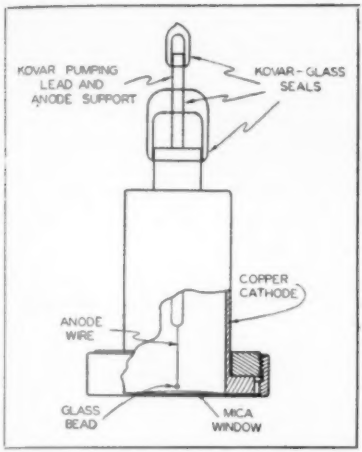

FIG. 5. Mica-window beta-ray counter

beta rays and still have sufficient mechanical strength.

Beta-ray counters designed with thin windows are, in general, quite different from the usual tube. A construction of this type of counter is shown in Fig. 5. The two main considerations which control the design are first, that the maximum number of beta rays enter the sensitive volume, and second, that the surface area of the cathode be small to minimize the counter's gamma-ray sensitivity. The background and gamma-ray sensitivity are reduced by decreasing the size of the counter, thereby reducing the surface area of the cathode. However, if the window area is to be kept large, there is a limit to the extent to which this can be done. Low gamma-ray sensitivity might be achieved by decreasing the length of the counter without changing the diameter. This, however, is not practical since a ratio of length to diameter of less than two gives too great a plateau slope and a very short working voltage range. To gain the advantage of a short counter without its poor characteristics, the usual center wire is replaced by one having a short section on the end with a small diameter, and the remainder with a larger diameter. This results in a counter whose beta-ray sensitivity is the same as though the center wire were of usual design, but whose gamma-ray sensitivity is greatly reduced. At the same time, the beta-ray plateau curve is as flat as that obtained with a normal anode wire.

The reason for the low gamma-ray sensitivity is as follows: Because of the smaller diameter of the center wire at the window end of the counter, this region reaches threshold for counting at considerably lower voltage than the central portion. Thus, the space in which an electron must appear in order to initiate a count is limited to a volume between the cathode and the short length of small-diameter center wire. Gamma-ray counts are obtained only when the gamma ray produces, at the cathode surface, secondary electrons which are ejected into the active volume of the counter. Therefore, all of the secondary electrons ejected are counted only from that part of the cathode surface which borders the active volume. For all other parts of the cathode, only those secondary electrons which are ejected in such a direction as to reach the active volume are counted. The contribution to gamma-ray sensitivity of a given area on the cathode depends on the solid angle subtended by the active volume at that point. The beta-ray sensitivity is not affected by these considerations, since the solid angle for a beta ray coming into the active volume from the window is independent of the depth of the counting volume.

In contrast with a short cathode counter, a counter with a cathode of normal length and a short stepped center wire has satisfactory plateau characteristics. The necessary large ratio of length to diameter is maintained, minimizing end effects which cause

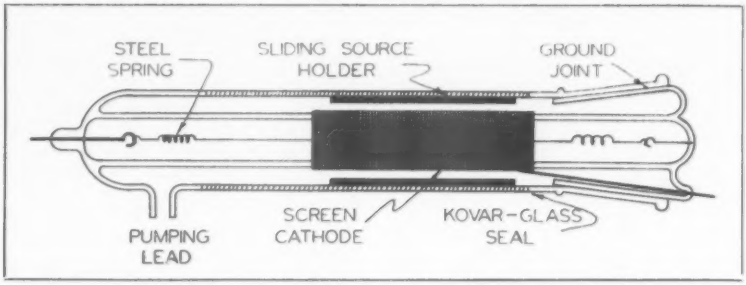

FIG. 6. Counter for low-energy beta rays; source may be placed inside envelope

poor characteristics; also, the gamma-ray sensitivity is equivalent to a counter of short cathode length.

The minimum length which can be used for the small diameter part of the center wire depends on the sensitivity of the amplifier connected to the counter. This is due to the fact that the size of the voltage pulse obtained from a counter is proportional to the effective length of the center wire. With the usual amplifiers, pulses coming from center-wire lengths below one centimeter are too small for satisfactory operation of the counter.

The principal advantages of this type of end-window counter are the large solid angle subtended by the sensitive counter volume at the window and the possibility of using very thin windows. With mica windows an inch in diameter, it is quite possible to support atmospheric pressure on a window 10 microns in thickness. Thinner windows may be used if pressure differentials are less, or if the window is supported on a metal screen.

The disadvantages of this type of counter are the more complicated construction and the distortion of the electric field caused by the presence of the window. At high counting rates, charges tend to collect on the inside of the window to such an extent as to change the characteristics of the counter. This effect may either give spurious counts or alter the electric field enough to change the effective voltage. One way of minimizing this effect is to coat the outer surface of the mica window with a thin layer of colloidal graphite.

For the detection of very soft beta rays, any windows between the source of radiation and the sensitive volume of the counter can be a serious disadvantage. Several counter arrangements have been used in which no window is necessary. The first such counter to be described was one by Libby and Lee (*5*).

This type of detector is shown in Fig. 6. The counter consists of the screen-walled cathode and fine-wire anode mounted axially in the cylindrical chamber. Just inside the outer envelope walls is mounted a cylindrical source holder which slides snugly inside the envelope. The source to be measured is stuck to the inside of this source holder. The sensitive volume of the counter occupies only the center section of the tube. Thus, the source may be slid in and out of the counting position by tipping up one end of the tube. Background counting rates are determined with the source holder pushed to one end of the tube, and the intensity of the sample is measured with the source centered over the screen cathode of the counter. The type and pressure of the filling gas can be anything con-

venient. It is found necessary to maintain a potential of a hundred volts or so between the counter cathode and the source holder and metal envelope. The direction of polarity of this sweeping potential is such that the source holder and envelope should be positive relative to the counter cathode. It serves the purpose of sweeping out ions which are formed outside the counter volume.

As one can see from Fig. 6, the source is changed through a large ground joint at the end of the counter. The disadvantage of this type of counter is the complication of changing sources, which involves disassembling at least part of the counter. The advantage, beyond the obvious one of no window, lies in the large solid angle for counting which the geometrical arrangement provides. Where samples of low specific activity must be measured, the large amount of material which may be placed in the counting position leads to high over-all efficiency.

Another arrangement for detecting low-energy beta rays (*6*) is shown in Fig. 7. The apparatus consists of a screen-cathode counter with a plunger, which holds the source, sliding in a side arm on the counter envelope. Helium from a commercial tank flows into the counter, down over the source, and out into the air through a bubbling bottle which acts as an air lock in case the helium is shut off. Helium, at approximately atmospheric pressure, flowing through the counter in this manner, is used for three reasons: At atmospheric pressure, it is not necessary to have any window between the source and counter, so that very low energy beta rays can be measured. If the helium is kept flowing gently through the apparatus at all times, the apparatus does not have to be made vacuum tight, leading to simplicity in construction and operation. Helium is used so that at atmospheric pressure the counter will operate at voltages which are easily obtainable

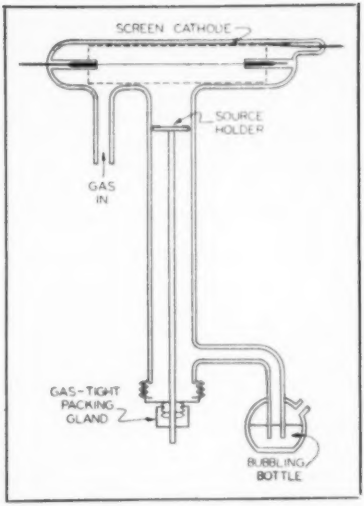

FIG. 7. A flowing-gas counter for detecting low-energy beta rays

by ordinary counter amplifiers. For example, such a counter, with a cathode diameter of two centimeters and anode of six mils, has a threshold voltage of 1,200 volts. If self-quenching action is desired, the input helium may be passed through alcohol. In this case, the threshold voltage wire will be a few hundred volts higher. Self-quenching gas, such as methane, which may be obtained commercially in tanks, also proves satisfactory.

The disadvantage of this type of low-energy beta-ray detection is the small size of the source which may conveniently be used on the sliding plunger. It has the advantage, however, of allowing quick and easy changes of source, and requires no evacuating and filling system.

Low-Background Systems

For counting low-intensity samples, a prime requisite is the minimizing of the background counting rate without decreasing the sensitivity for detecting

the sample radiation. The simplest method of achieving this result is to surround the counter with as thick a lead shield as possible, including the source within the lead shield. This procedure is often inconvenient or impossible, and various methods have been tried for obtaining low-background counting systems without bulk shielding.

Background radiation usually consists of gamma rays, or cosmic-ray secondaries, which are most efficiently detected by the gamma-ray type of counter. The construction of the step-center-wire beta-ray counter is advantageous because the gamma-ray sensitivity is low. The small cathode surface means low gamma-ray efficiency, but the beta-ray efficiency is close to 100 percent because of the small probability of an electron's traversing the sensitive volume without creating a breakdown. Beta-ray counters built with thin glass walls to allow the entrance of the beta rays are inefficient gamma-ray counters, since the walls are poor secondary emitters due both to their thinness and to the low atomic weight of glass.

The types of counter construction which lead to directional properties can be effectively used to reduce background. For example, the beaded center-wire, self-quenching type of counter can be made very efficient in this regard. If an end-window, beta-ray counter is made with a beaded axial wire, the beta rays which enter parallel to the counter axis will cause discharges in all the sections, whereas gamma rays or other radiation entering from the side will cause a discharge in one section only and hence, can be distinguished easily.

The coincidence methods discussed in the section on directional counters can also be used for reducing background counting rate, since the source can be placed in the most advantageous geometry for producing the coincidence count.

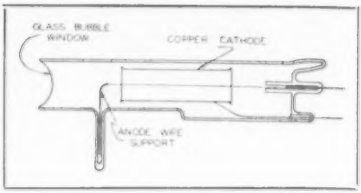

FIG. 8. A bubble window counter

X-RAY COUNTERS

The problem of detecting X-rays is much the same as for beta rays. For high efficiency, a window must be provided to allow the radiation to reach the sensitive volume with a minimum of attenuation. Thin glass windows in the form of a bubble are often used.

In detecting radiation from an X-ray tube, one is usually dealing with a collimated beam and, for this reason, there is no necessity of having the sensitive volume close to the window. This allows a construction in which both ends of the center wire are supported, providing considerably more rigidity than is possible with the usual end-window type of counter. The construction of such a counter is illustrated in Fig. 8.

Special glasses, such as Lindemann glass (3%, BeO; 81%, $Li_2B_4O_7$; 16%, B_2O_3), are sometimes used where the decrease in window absorption warrants the use of special techniques and glasses. Beryllium has a high transmission for X-rays and may be used as a window in the same manner as the mica-window counter illustrated in Fig. 5. Beryllium has certain advantages; for example, by making the window 0.04 inches thick, an X-ray counter can be rendered insensitive to beta rays up to 0.5 Mev while maintaining a high efficiency for detecting X-rays. This results from the fact that the beta rays cannot penetrate this thickness of beryllium while X-rays will.

OPTICAL PHOTON COUNTERS

The cathode of a counter can very easily be built of a photosensitive material, and, since the counter is capable of detecting every singly emitted photoelectron, it can be made into a very sensitive photon detector. The difficulties, however, in producing stable and reproducible surfaces are great. The principal reasons are: first, the cathode surface is subject to intense ion bombardment in the process of the discharge and, hence, must be unusually stable; second, photosensitive surfaces are subject to both contamination and/or activation by the gas and gas products in the discharge; and third, almost nothing is known theoretically about the photosensitive characteristics of surfaces in the presence of the gases used in counters.

One can say, in general, that the photosensitivity of cathodes in a gas parallels that in a vacuum, and, therefore, one can make a reasonable guess at the spectral sensitivity characteristics by examining the data obtained in a vacuum. A good deal of systematic work has been done on this problem (*7*), but thus far it is more probable to find high photoelectric sensitivity by chance coincidence of uncontrolled factors than to approach the uniformity of vacuum photoelectric devices.

The studies which have been made on the photosensitivity of counter cathodes have shown that the peak sensitivity lies in the 2,500 A.U. region. For metal cathodes, such as aluminum, zinc, and copper, treatments such as activation in a hydrogen discharge increase the sensitivity (*8*). Coating the cathodes with films of calcium, magnesium, or sodium has also been shown to increase the photosensitivity (*9*). Activation of the cathode by such unpredictable treatments as argon-butane gas discharges at liquid air temperatures increases photoelectric sensitivity (*10*). Much more work on this problem must be done before any general rules or theories can be set down to guide the experimenter in the field of optical photon counters.

COUNTERS FOR LIQUID SOURCES

The principal problem in counting a radioactive liquid sample lies in holding the liquid in a sufficiently rigid geometrical arrangement to obtain reproducible results. For beta rays, this is relatively simple. The internal absorption of the source is so high that small thicknesses of source liquid become an infinitely thick source as far as the beta rays are concerned, and beyond this, the volume does not make any difference. In this case, the problem is the usual beta-ray detection problem of designing thin enough counter walls to minimize the attenuation of this radiation. Figure 9 shows a successful design of this type. The thin, thimble-shaped end has a deposited metal coating on the inside surface to form the cathode. Since all of the electrical connections are led through one end, well-removed from the sensitive volume, the counter may conveniently be dipped into the liquid to be measured.

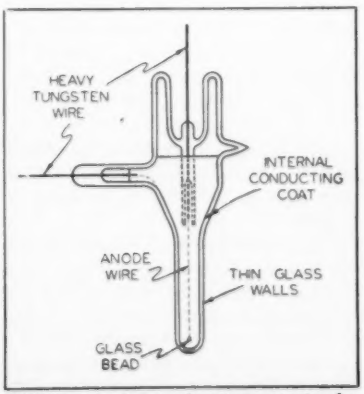

FIG. 9. An immersion-type counter for detecting liquid sources

Unless great care is exercised in holding a rigid geometry, dipping counters are not so useful for measuring any radiation where the thickness of the liquid changes the counting rate, as for soft radiation where the source can be infinitely thick.

For counting gamma rays efficiently from a liquid, an ordinary gamma-ray counter surrounded by a glass jacket for holding the liquid can be used. If this is built in one piece, there is no chance of geometrical changes in counting routine samples, provided the jacket is always maintained full of the radiating liquid. Such a counter is illustrated in Fig. 10. Special cells for holding liquid samples for use with end-window type of beta ray counters have been described in the literature (*11*).

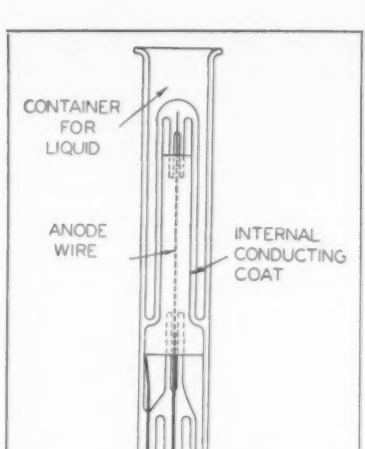

FIG. 10. A gamma-ray counter for liquid sources

COUNTERS FOR GASEOUS SOURCES

Since the filling gas of a counter controls the discharge mechanism, the problem of using special gases presents unique problems for each gas. In the first paper of this review series, the discharge mechanisms were discussed at length. When the gas in a counter must be specified for some experimental reason, knowledge of the gas characteristics will indicate possible solutions. As a case in point, let us choose as a particular example a counter filled with carbon dioxide (*12*). This gas is useful in tracer experiments using the long-lived carbon isotope, since the radioactive carbon can be converted by relatively simple chemical procedure into this gaseous compound in which the weight of the carbon in the molecule is great.

Carbon dioxide is not a good gas to use in a counter because it forms negative ions. Reference to Table 1 of the previous paper of this series shows that although it does not form negative ions by electron attachment, it does form them in collision with the walls. It is necessary to minimize this negative ion formation if successful counter operation is to be achieved. As far as is known, it is impossible to eliminate negative ion formation at the cathode from the CO_2 ions. Therefore, these ions must be prevented from reaching the cathode.

After the discharge is initiated by an entering ionizing particle, cumulative ionization creates a positive-ion sheath around the central wire anode. This positive-ion space-charge sheath is repelled by the anode, around which it was formed, and is attracted toward the cathode. The mobilities of the positive ions are such that it takes of the order of 10^{-4} seconds to travel across a one-centimeter electrode gap, and in this time the ions may make as many as 10^4 collisions with neighboring gas molecules. If another gas whose ionization potential is less than that of the main gas is mixed with the CO_2, there will be a high probability that CO_2 will collide

with the other type of gas molecule, ionizing it at the expense of the charge on the carbon dioxide ion. This is the charge transfer mechanism previously discussed. There are quite a number of gases which may be used to transfer the charge from the carbon dioxide ion. Carbon disulfide is a particularly good one.

In some problems in the use of counters for special purposes, difficulty may arise from the necessary choice of filling gas, but there may be nothing that can be done to alter the discharge mechanism for satisfactory operation. For example, in the study of respiratory problems (*13*), Geiger-Müller counters show the following difficulties: The threshold voltage for air at atmospheric pressure is very high. Exhaled air contains a large percentage of carbon dioxide and water vapor. If the experiment is to be carried out in a closed system, the relative concentrations of nitrogen and oxygen change, resulting in a change in operating voltage.

These difficulties can be overcome in the following way: The CO_2 can be reduced below significant concentrations by means of a soda-lime trap. The principal trouble with the presence of water vapor is that it may condense on the envelope of the counter and form a direct leakage path for the high potential. This may be prevented by heating the counter, and always operating it at a temperature above 100° C.

Changes in threshold voltage with gas composition are equivalent to changes in voltage applied to the counter. It is thus possible to introduce a second counter, working on the same characteristics, to feed back a voltage to compensate for the change in threshold voltage caused by gas composition. This second counter is connected to the first so as to share the same gas. The second counter differs from the first only in that it contains a source of Ra-D, which emits beta rays but no gamma rays. This counter could be called the control counter whereas the first is the measuring counter.

The procedure is to operate the control counter slightly above threshold where counting rate changes rapidly with changes in voltage. This change in counting rate is converted to a change in voltage by an electrical integrating circuit, and applied as a reversed potential to the high-voltage supply. Any increase in counting rate in the control counter thus produces a decrease in voltage applied to both counters. The voltage from the stabilized, high-voltage supply is raised until the control counter reaches threshold. If the measuring counter is set a fixed voltage above its threshold, the correction voltage circuit will maintain it at this point independent of changes in gas concentration and stabilized high voltage changes.

SPECIAL COUNTERS

Unusual Size

Counters of any geometrical shape can be made to operate although, in general, counters of odd shapes serve no very useful purpose. Their sensitivity, in general, is greatly reduced. The occasion does sometimes arise when it is useful to make counters of the usual cylindrical form but either very large or very small.

Counters of very large diameter have two characteristics which must be recognized. First, they are usually very slow since the ions have a long path length and cannot be cleared rapidly from the sensitive volume. In general, to avoid excessively high operating voltages, the center wire is kept small in diameter, and only the cathode size is increased. This brings the multiplying field relatively close to the anode, causing essentially a field-free space extending inward from the

cathode for some distance. In this very weak field region, ions may become trapped for relatively long times, producing spurious discharges when they drift out of the ionizing region. This effect can assume serious proportions in very large counters. Second, very large counters have very high background rates due to the large surface area for secondary production from gamma rays or cosmic rays. Unless this is the desired result, their usefulness is seriously impaired by this background. Very long counters have the disadvantage of a high capacitance, which increases the electrical time constant and, therefore, makes the counter response slow.

Very small counters have many uses, both in physical measurements and applied radioactivity studies. They have not, so far, proved as satisfactory as the more conventional sizes. Although the electrodes can be decreased to almost any size that it is possible to build, the glass ends and seals cannot be scaled down in the same way for practical constructional reasons. This leads to a decrease in the relative size of the sensitive volume compared with the total volume of the counter, and, hence, the loss in over-all efficiency is greater than one expects from the simple change in size. Small counters are also subject to a hysteresis effect, which, after exposure of the counter to a high radiation intensity, prevents the counting rate from returning immediately to its background value on removal of the source. This effect may be a result of the collection of charges on the relatively large glass ends.

Very small counters often show a greatly decreased length of life. When the counters are filled with polyatomic gas fillings, this is understandable on the basis of the smaller total number of molecules present which must be dissociated in the discharge as an essential part of the counting mechanism.

Large Solid-Angle Counters

With the conventional counter, in which the source is always outside the sensitive counter volume, the maximum, theoretical, solid angle which the source can subtend is 2π. A number of attempts have been made to design Geiger-Müller counters for greater solid angles, and these are characterized by different methods of introducing the radioactive source into the sensitive volume of the counter.

Brown and Evans (*14*) undertook a fairly extensive study of the problem, with particular attention being given to the behavior of counters of unconventional geometry. The results of their work showed that essentially any geometry could be made to function, although most forms were too insensitive to be practical. The most successful large solid-angle counters of this type were coaxial cylinders in which the source could be placed within the axial electrode. These counters are inherently slow compared with the standard Geiger-Müller geometry, since the more homogeneous field does not restrict the ion multiplication to the vicinity of the anode. It, therefore, takes longer for the electrons to be collected. This is not a serious disadvantage since the use of high solid-angle counters is not warranted except with weak sources. The serious disadvantage of these large coaxial anodes is a loss in sensitivity which, in most cases, nullifies the gain in solid angle.

It should be pointed out that, if large coaxial cylinders are to operate as Geiger-Müller counters, there is a minimum ratio of cathode to anode radii below which a corona discharge cannot exist. This limit is the Naperian logarithm base, 2.718. To recognize this, one must realize that the corona formed by the ionized atoms or molecules of the gas forms a conducting sheath around the inner conductor. This sheath increases the effective

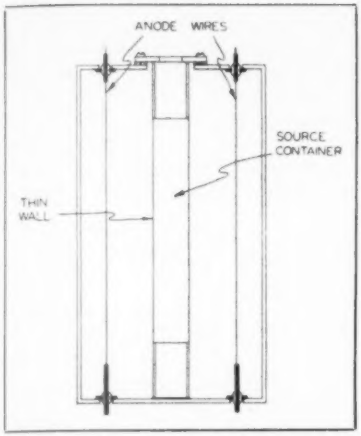

FIG. 11. A large solid-angle counter in which the source may be placed in a thin-walled box mounted between two anode wires

radius of the anode. If this increase lowers the field, the conditions for an ordinary corona discharge are fulfilled. If, on the other hand, the increased radius raises the field, a disruptive discharge will take place from anode to cathode and the corona region will be absent. One can show that the field is increased if $r_c/r_a < e$, but is decreased if $r_c/r_a > e$, where e is the Naperian logarithm base.

To avoid the loss in sensitivity which occurs when one deviates from the small anode wire, several types of multiple anode-wire counters have been suggested. High solid-angle counting can be achieved by surrounding the source with the sensitive volume of two or more center wires. Simpson has described two such counters in detail (*15*). One of these consists of a single cylindrical cathode in which is mounted two off-axial anodes. A thin metal box, the inside of which is open to the air, is supported between these anodes. Thus, a source, mounted in the metal box, will subtend a large solid angle with the combined sensitive volumes of both anode wires. This type of construction is illustrated in Fig. 11. The other type which Simpson describes is essentially two ordinary Geiger-Müller counters with the cathodes flattened on one side. The two flat sides close together with the source clamped between.

Maximum solid angle is always achieved if the source can be introduced as a filling gas. From this point of view, counting low intensity sources in the gaseous phase is always preferable. For the special problems involved, see the section on special gases on page 60.

Low-Voltage Counters

The operating voltage which is required for a counter is given by three variables: the electric field, the pressure, and the Townsend ionization coefficient which is a measure of the number of ion pairs formed per volt travel of an electron in the field. Attempts have been made to produce low-voltage counters by controlling each variable.

One can adjust the operating voltage of a counter of given gas filling and geometry by changing the filling pressure. There is a limit, however, to the amount one can lower the pressure without seriously decreasing the efficiency of detection. It is found that for most gases, if the pressure is reduced below about six centimeters of mercury pressure, the probability for a single electron to trigger a discharge decreases rapidly. For counters of usual geometry, when the pressure has been reduced to one half a centimeter of mercury the mean free path of the electron approaches the distance of travel between the cathode and anode and the breakdown voltage goes through a minimum, due to the fact that ionization efficiency is so low that the field must be increased to cause breakdown. One is limited, therefore, to minimum threshold voltages of around 1,000 volts for ordinary gases because of the loss of efficiency at low pressures.

Since the threshold voltage for a given gas depends on the energy that the electrons forming the avalanche pick up per mean free path, this voltage depends directly on the electric field. The electric field is given by the relation:

$$E = \frac{V}{r \ln r_c/r_a},$$

so that if one keeps the radius of the anode, r_a, constant and decreases the cathode radius, r_c, one can obtain a higher field at a lower voltage. However, to get a significant reduction in operating voltage, the counters become so small that their over-all detection efficiency is seriously impaired. Korff and Ramsey have suggested the use of a negative grid close to the anode to act as the field cathode (*16*). If the ordinary counter cathode is made slightly negative with respect to this grid, the detection efficiency is equivalent to a gamma-ray counter with a cathode of usual diameter. The construction of such a grid counter is difficult and the reduction in threshold is about 30 percent.

The most satisfactory way of obtaining low-voltage operation for Geiger-Müller counters is to choose a suitable gas filling. It was pointed out in the first section of this review series that the Townsend ionization coefficient had particularly high values for low field strengths for neon-argon mixtures. The neon atom has a metastable level at 16.5 volts which is above the ionization potential of argon (15.7 volts). Probabilities of excitation are in general high compared to ionization probabilities. Furthermore, since, in a collision between a metastable atom and one whose ionization potential is low, the metastable atom will almost always ionize the second atom, one can consider that essentially all neon excitations result in ionizations (of the argon). This ionization can very easily be produced in neon with traces of argon added. The actual ionization coefficients for these mixtures, have been measured by Kruithof and Penning (*17*) and their characteristics as counter gases have been measured by Simpson (*18*). These filling mixtures lead to an operating voltage of only a few hundred volts without loss of any of the desirable Geiger-Müller characteristics. Because of the extreme purity of the gases required before the low-voltage characteristics can be realized, ordinary counter filling technique cannot be used. Only very good high-vacuum technique and great care in handling and purifying the gases will produce the low-voltage conditions.

In this article, we have considered the construction and behavior of a number of counters for specific purposes without discussing any of the electronic circuits which in some cases are an integral part of their successful operation. The electronic equipment will be described in the next article of this series.

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NUCLEONIC EVENTS

CANCER GROUP TO GIVE GRANTS, FELLOWSHIPS

The Committee on Growth of the National Research Council, acting for the American Cancer Society, has announced that it is accepting applications for grants and fellowships. Applications for extension of existing grants in cancer research will be received until October 1, applications for new grants until November 1. Final decision on applications submitted during this period will be made, in most cases, soon after February 1. Grants approved at this time ordinarily will become effective July 1, 1949.

Fellowship applications may be submitted at any time. Those received prior to November 1 will be acted upon by the Committee on Growth in December. Those received between November 1 and March 1 will be acted upon in April. Fellowships ordinarily will begin July 1 though this date may be varied at the request of the applicant.

Communications regarding grants and fellowships should be addressed to Executive Secretary, Committee on Growth, National Research Council, 2101 Constitution Avenue, N. W., Washington 25, D. C.

"NUCLEONICS AND ANALYTICAL CHEMISTRY" SYMPOSIUM SUBJECT

The symposium on nucleonics and analytical chemistry held by the American Chemical Society at Northwestern University this month included the following papers: Instruments Employed in the Measurements of Radioactivity by C. J. Borkowski, Techniques Employed in Applied Radiochemistry by W. H. Sullivan, Radioactive Isotopes as Tracers by P. Yankwich, Determination of Natural Occurring Radioactive Elements by C. J. Rodden, Activity Analysis by D. N. Hume, Activation Analysis by G. E. Boyd, Industrial Applications by J. W. Irvine, Jr., and Preparation and Procurement of Radioactive Materials by N. H. Woodruff and S. A. Lough.

IRE, AIEE TO HOLD CONFERENCE ON NUCLEONICS INSTRUMENTS

A tentative program covering 19 subjects has been set up for the Joint IRE-AIEE Conference on Electronic Instrumentation in Nucleonics and Medicine to be held in New York City on November 29, 30, and December 1.

Scheduled for the first day, with W. A. Geohegan of the Cornell University Medical College as chairman, are papers on the following subjects: biological requirements in amplifiers; present practice in biological amplifier design; biological requirements in recording devices, including cathode-ray oscillograph, electrocardiograph, and the electroencephalograph; and present practice in biological recorder design.

G. W. Dunlap of the General Electric Company will be chairman of the second day's program in which the following will be discussed: biological requirements for radioactive isotope measurements; stable isotope measurement; thin-window beta counters; electron multiplier counters; crystal counters; health protection instrumentation; and health protection techniques.

The final day will have H. H. Goldsmith of Brookhaven National Laboratory as chairman. The program will include papers on: geiger counters,

proportional counters, including alpha and neutron (BF_3 and boron-coated); ionization chambers and ionization chamber measurements; stabilized high-voltage supply for counters and chambers; cloud chambers; photographic emulsions; high-speed counting techniques; and autoradiography.

AEC ESTABLISHES NEW CHEMICAL LABORATORY

The Atomic Energy Commission will establish a chemical laboratory in New Brunswick, New Jersey, to provide precise analytical quality control of special materials, according to a recent announcement by W. E. Kelley, manager of New York Directed Operations of the AEC.

To be known as the New Brunswick Laboratory, the new AEC installation, which will begin operation in the fall, will provide facilities for quality control of material processed in plants under the supervision of the AEC's New York office. Operations under this office include the processing of uranium, beryllium and other ores and materials.

TWO SOVIET SCIENTISTS DISCOVER NEW PARTICLES

The discovery of fourteen new subatomic particles, identified as "varitrons," in cosmic rays has won for two Armenian scientists, A. I. Alikhanyan and A. I. Alikhanov, one of the Soviet awards for prominent scientific advances during 1947.

The discovery was made during the course of cosmic ray studies that the Armenian nuclear physicists have been conducting every summer and autumn since 1942 at a research laboratory established at an elevation of 1.9 miles above sea level on Mount Alagez in Soviet Armenia.

The Alikhanovs have discovered new particles with masses of 100, 150, 250, 300, 430, 550, 680, 1000, 1300, 2500, 3800, 8000 and 25,000 electron masses.

GENERAL ELECTRIC ISSUES ISOTOPE CHART

A new chart giving the latest data concerning the various known isotopes is being distributed by the General Electric Research Laboratory in Schenectady, New York. The chart, which was prepared by G. Friedlander and M. L. Perlman of GE, follows the general pattern of one previously devised by E. Segré of the University of California. Printed on heavy paper, 26 by 50 inches, it is in the form of a long diagonal checkerboard, divided into three overlapping sections to get it on a sheet of convenient size.

Copies of the chart, and a booklet describing it, are being distributed gratis to college departments of physics and chemistry as well as to other laboratories. Any person interested may obtain one free on request to GE, Schenectady 5, New York.

BETHLEHEM STEEL JOINS U. OF CHICAGO PROGRAM

The Bethlehem Steel Company has become the thirteenth industrial organization to join the University of Chicago's industry-education atomic research program. In investing $100,000, Bethlehem has joined the Institute of Metals. The Institute of Nuclear Studies and the Institute of Radiobiology and Biophysics are the other two groups in the program.

MORSE RESIGNS AS HEAD OF BROOKHAVEN LAB

Philip M. Morse resigned last month as director of Brookhaven National Laboratory, Upton, N. Y., according to an announcement by Associated Universities, Inc. Dr. Morse plans to return to his position as professor of physics at MIT.

Leland J. Haworth, former assistant director at the laboratory, has been appointed acting director.

ABSTRACTS

CHEMICAL PUBLICATIONS

A positive ion source, A. J. Bayly, A. G. Ward (Chalk River Lab., Natl. Research Council of Canada, Ont.), *Can. J. Research* **26A,** 69–78 (1948). An electrodeless discharge is used to produce the positive ions, which are then extracted from the discharge through a pumping canal by a suitable arrangement of electrodes. The beam consists of 50% protons when using hydrogen in the discharge and 60% deuterons when using deuterium.

Gamma rays produced in the fission of U^{235}, B. B. Kinsey, R. C. Hanna, D. Van Patter (Chalk River Lab., Natl. Research Council of Canada, Ont.), *Can. J. Research* **26A,** 79–98 (1948). Two radiations with an energy of 2.5 Mev are emitted in each fission process of U^{235}, but the sensitivity of the apparatus was not high enough to verify (by observing γ-γ coincidences) that two such γ-rays are emitted per fission.

Angular distribution of neutrons inside a scattering and absorbing medium, P. R. Wallace (McGill Univ., Montreal) *Can. J. Research* **26A,** 99–114 (1948). The moments of the angular distribution of monoenergetic neutrons diffusing inside a homogeneous medium were determined using the transport theory. A general derivation involving no assumptions about the relative scattering and capture cross sections is given for the current-density relation.

The detection of free radicals in hydrogen atom reactions with organic molecules, G. M. Harris, A. W. Tickner (Univ. of Melbourne and Univ. of Toronto), *Can. J. Research* **26B,** 343–355 (1948). A method is described for the preparation of radioactive bismuth mirrors of known specific activity using Bi^{210} obtained from uranium extraction residues. Such bismuth mirrors were used to study the reactions of hydrogen atoms with acetone and dimethyl mercury.

Production of radioactive carbon monoxide and phosgene from barium carbonate, J. L. Huston, T. H. Norris (Univ. of California, Berkeley), *J. Am. Chem. Soc.* **70,** 1969 (1948). Radioactive carbon monoxide can be made from $BaC^{14}O_3$ by decomposing the latter with phosphoric acid and reducing the C^*O_2 to C^*O with hot zinc. The procedure is quantitative and involves no dilution of radioactive carbon. Radioactive phosgene was prepared photochemically by the reaction of C^*O with chlorine, in yields of 80.0 to 84.5% based on the BaC^*O_3.

Radiation chemistry, V. Effect of molecular size, M. Burton (Univ. of Notre Dame, South Bend, Ind.), *J. Phys. Colloid Chem.* **52,** 810–818 (1948). With large molecules the amount of a product of a particular type may exceed by far the amount to be expected on the purely statistical basis. The explanation is that in large molecules, ultimate-molecule decompositions become more and more favored and occur preferentially over the lowest energy pass. The excitation energy may be distributed in a greater number of modes of excitation for large molecules, thereby increasing the probability of deactivation and favoring decomposition via a low-energy pass requiring a minimum of rearrangement.

The disintegration of antimony-124, I. Feister, L. F. Curtiss (Natl. Bureau of Standards) *J. Research Natl. Bur. Standards* **40,** 315–319 (1948). The spectrum of 60-day half-life Sb^{124} was investigated using a magnetic-lens electron spectrometer. The β-ray spectrum has two components of maximum energy 0.57 ± 0.02 and 2.24 ± 0.05 Mev, while the γ-ray spectrum has two lines, 0.60 ± 0.01 Mev and 1.69 ± 0.02 Mev. These measurements substantiate the disintegration scheme of Meyerhof and Sharff-Goldhaber.

Extensive penetrating showers, B. Chaudhuri (Univ. of Manchester), *Nature* **161,** 680 (1948). Showers selected by a fourfold array of G-M counters were used to trigger a cloud chamber. The operation of a large counter tray, heavily shielded by lead and connected in coincidence with the fourfold array, was recorded on the chamber photographs. The average probability that a single shower particle is accompanied by a penetrating event was found to be 0.020 ± 0.002. The expected rate of photographs showing a number n of shower particles, and the fraction f of photographs showing n tracks not accompanied by penetrating counts were calcu-

lated and compared with experimental results.

Mesons of different masses, J. Barnóthy (Univ. of Budapest), *Nature* **161,** 681 (1948). A meson theory is proposed which involves the existence of electrons and protons in unstable "meso-states" as well as in their normal stable state. This theory gives one solution for the meso-state of the electron and eighty-two solutions for the meso-protons. It is shown how the experimental facts concerning mesons of different masses can be explained by this theory.

Scattering of 100-Mev neutrons by protons, F. C. Barker (Univ. of Birmingham), *Nature* **161,** 726–727 (1948). Using an exponential-well potential $V_0e^{-r/a}$ with two values of the range a, calculations were made for the differential cross section for the scattering of 100-Mev neutrons by protons. The two cases of exchange and symmetric forces were assumed. The type of force has little effect on the shape of the differential cross section.

Comparison of cross sections for the capture of 220 and 900 kev neutrons, R. Allen, G. R. Bishop, P. Demers, H. Halban (Oxford Univ. and Univ. of Montreal), *Nature* **161,** 727 (1948). The ratio of the capture cross sections of twenty-three nuclei for neutrons of 220 and 900 kev were determined. The ratios were obtained by irradiating the same sample of each element in turn with each of the two neutron sources, and then measuring the ratio of the intensity of artificial radioactivity obtained in the two irradiations. The ratios $\sigma 220/\sigma 900$ vary between 1 and 4.

Emission of Li^8 in the explosive disintegration of nuclei, C. Franzinetti, R. M. Payne (Univ. of Bristol), *Nature* **161,** 735–736 (1948). Photographic plates showed twenty-eight examples of the emission of a nucleus of Li^8 during an explosive disintegration, followed by decomposition of the Li^8 into two α-particles of equal range, but emitted in opposite directions. One photograph provides evidence for the ejection of Li^8 in the disintegrations produced by mesons.

Angular distribution of radioactive disintegration products, J. A. Spiers (Oxford Univ.), *Nature* **161,** 807–809 (1948). Theoretical calculations have been made on the angular distribution of radioactive disintegration products originating from a partially oriented nuclear-spin system. Such partial orientation can be obtained by the application of high magnetic fields at temperatures of about 0.01° K, followed by adiabatic demagnetization.

Capture of negative mesons by nuclei, A. S. Lodge (Oxford Univ.), *Nature* **161,** 809–810 (1948). A hypothesis is proposed to explain the unexpectedly long period which has been found for the nuclear capture of negative mesons from cosmic rays at low altitudes. It is assumed that π-mesons alone have direct coupling with nucleons, and a scalar wave function is taken to describe the π-meson field. There is an indirect interaction between light mesons and nucleons by virtue of the coupling implied in the disintegration of heavy mesons into light mesons.

Cloud chamber investigation of straggling of short-range particles produced by slow neutron reactions, J. K. Boggild (Inst. for Theoret. Physics, Copenhagen), *Nature* **161,** 810–811 (1948). The relative straggling coefficient of protons was found to be 0.020, in good agreement with theory. The range distribution for the process $_5B^{10} + {}_0n^1 \rightarrow {}_3Li^7 + {}_2He^4$ could not be represented by a simple Gaussian curve, indicating that nuclear collisions contribute appreciably to the straggling effect.

. . . I. W. RUDERMAN

LIFE SCIENCE PUBLICATIONS

Effects of radioactive sodium on leukemia and allied diseases, T. Evans, M. Lenz, C. Donlan, M. LeMay (Dept. of Radiology, Columbia Univ., New York), *Am. J. Roentgenol. Radium Therapy* **59,** 469–481 (1948). This is a preliminary report permitting only tentative conclusions. Radiosodium administered orally in suitable quantities and at appropriate intervals was found effective in reducing symptoms of chronic lymphatic leukemia, polycythemia vera and chronic myelogenous leukemia. Contra-indications are similar to other forms of radiation therapy.

Experimental therapy of onchocerciasis with trivalent antimonials, F. Bartter, T. Burch, D. Cowie, L. Ashburn, F. Brady (Div. of Tropical Diseases, National Inst. of Health, Bethesda, Md.), *Ann. N. Y. Acad. Sci.* **50,** 89–96

(1948). Patients with onchocerciasis were treated with stibophen and tartar emetic. Posttreatment examination of skin and nodule biopsies did not reveal a significant decrease in the numbers of microfilariae or in the motility of adult parasites. The tartar emetic was prepared with radioactive antimony permitting the measurement of the antimony uptake by adult parasites present in a nodule. This uptake was similar to that of adult Dirofilaria immitis obtained from a dog successfully treated for filariasis. There is some evidence that the adult onchocerca females were injured by the therapy.

The span of life of the red blood cell. A resume, W. Ashby (St. Elizabeth's Hospital, Washington, D. C.), *Blood* **3,** 486–500 (1948). A review of experiments performed involving various techniques to determine the life span of the red blood cell. Several different methods agree to an average life span of 110 to 130 days under favorable conditions. The author's studies of transfused Group O blood are reviewed in some detail.

Studies in iron transportation and metabolism—VI. Absorption of radioactive iron in patients with fever and with anemias of varied etiology, R. Dubach, S. Callender, C. Moore (Dept. of Internal Medicine, Washington Univ., St. Louis, Mo.), *Blood* **3,** 526–539 (1948). It was shown that patients with fever, untreated pernicious anemia, and refractory anemia absorb more iron than they use for hemoglobin. It is suggested that the theory that mucosal cells accept iron for absorption or block its assimilation provides the best known explanation for iron absorption; but patients with adequate iron stores may assimilate considerable quantities of the metal, and the block must be regarded as relative. The recovery of radioiron from feces was shown accurate within 10%.

Some hematologic effects of irradiation, W. Bloom, L. Jacobson, (Inst. of Radiobiology and Biophysics, Univ. of Chicago, Ill.), *Blood* **3,** 586–592 (1948). Studies involving the acute and chronic application of externally originating ionizing radiations and internally deposited radioisotopes gave no evidence of primary stimulation of hematopoiesis. Secondary increases in some of the cellular constituents of the peripheral blood were observed and were invariably preceded by a reduction. An initial leukocytosis which occurred in the first 24 hours after acute exposure to externally originating irradiations is probably a reaction to injury not involving a new formation of blood cells.

The effect of radiation on hemopoiesis. Is there an indirect effect?, J. Lawrence, W. Valentine, A. Dowdy (Dept. of Medicine and Radiology, Univ. of Rochester, N. Y.), *Blood* **3,** 593–609 (1948). Carotid to carotid anastomoses were performed between normal cats and radiated cats. Cross circulation was established in most cases at some specified time interval after the radiation of one partner. Leukocyte and lymphocyte counts in the normal animals observed during an approximate 28-day period do not lend support to the thesis of indirect effects peculiar to radiation.

Studies in intermediate metabolism conducted with the aid of isotopic tracers, D. Stetten, Jr., (Dept. of Biological Chem., Harvard Med. School, Boston, Mass.), *Bull. N. Y. Acad. Med.* **24,** 87–96 (1948). A review of the use of isotopes as a method of studying intermediate metabolism.

The effect of radioactive phosphorus upon a suspension of escherichia coli, C. Schmidt (Research Dept., Continental Can Company, Chicago, Ill.), *J. Bact.* **55,** 705–709 (1948). The addition of radiophosphorus to a suspension of Escherichia coli in phosphate buffer had a lethal effect upon the cells. However, complete sterilization of the suspension was not obtained even in the highest concentration tested. The survival curve indicated that some cells are more resistant to radiation than others. The cost of using radioactive phosphorus for the sterilization of food products would be prohibitive. As a result, no possibility of the use of radioactive materials for food sterilization is suggested.

Changes in phosphate and carbohydrate metabolism in shock, E. Giranson, J. Hamilton, R. Haist (Dept. of Physiology, Univ. of Toronto, Canada), *J. Biol. Chem.* **174,** 1–9(1948). Uninjured muscle of rats shocked by the clamping technique suffered a glycogen concentration decrease which was not prevented by insulin administration. Phosphocreatine (PC) and ATP content of damaged muscle fell because of clamping procedure and remained low. After clamping, the inorganic P level fell but was still above normal. PC and ATP content of undamaged muscle of shocked

animals was the same as the non-clamped controls. The P^{32} values for PC, ATP and inorganic P was elevated by clamping. Insulin increased incorporation of P^{32} in PC and ATP in normal fasted controls. Blood inorganic phosphate level rose markedly in shocked rats and was associated with a reduction of inorganic P in the damaged muscle after removal of clamps. Depletion of high energy P is not believed an initiating factor in shock.

The distribution of potassium isotopes in biological material, L. Mullins, K. Zerahn (Inst. for Theoretical Physics, Univ. of Copenhagen, Denmark), *J. Biol. Chem.* **174,** 107–113(1948). The isotopic K^{40} content of animal, mineral and vegetable sources were compared with reagent KCl. No significant difference in concentration was observed.

Distribution studies—XI. Isolation of benzylpenicillin containing radioactive sulfur, Y. Sato, G. Barry, L. Craig (Labs. of the Rockefeller Inst. for Med. Research, N. Y.), *J. Biol. Chem.* **174,** 217–220(1948). Labeled amorphous crude penicillin was obtained from Penicillium notatum grown in medium containing radioactive sulfur as sodium sulfate by means of counter-current distribution process. Labeled crystalline benzylpenicillin was readily isolated.

A detailed procedure for the preparation of highly purified adenosine triphosphate, A. Dounce, A. Rothstein, G. Beyer, R. Meier, R. Freer (Depts. of Biochemistry and Radiology, Univ. of Rochester, N. Y.) *J. Biol. Chem.* **174,** 361–370 (1948). A preparation of ATP containing radiophosphate is described.

The tissue distribution of radioantimony inhaled as stibine, R. Smith, J. Steele, R. Eakin, D. Cowie (Naval Medical Research Inst., Bethesda, Md.), *J. Lab. Clin. Med.* **33,** 635–643 (1948). Radioactive antimony in the form of stibine gas, SbH_3, was administered to normal guinea pigs, normal chicks and chicks infected with P. gallinaceum. The amount of radioactive antimony present in blood and tissues at successive time intervals was measured. There was no significant difference in distribution found between the normal and infected groups.

The red cells initially contained a much higher concentration than did the plasma, but the difference fell with time. Similar concentration curves of antimony were obtained for lung, brain, muscle, fat and blood. These were smoothly decaying curves. The curves for liver and spleen passed through a maximum about one hour following treatment. Evidence is presented that the rate of elimination of antimony from the body is higher for the guinea pig than for the chick.

Radiosodium studies in heart failure, *Lancet* **254,** 527(1948). A brief review of an experiment involving the use of radiosodium, Na^{22} and Na^{24} in studying chronic congestive heart failure.

Treatment of euthyroid cardiac patients by producing myxedema with radioactive iodine, H. Blumgart, A. Freedberg, R. Buka (Dept. of Medicine, Harvard Med. School, Boston, Mass.), *Proc. Soc. Exptl. Biol. Med.* **67,** 190–191 (1948). Two severely ill cardiac patients were given radioactive iodine, I^{131}, but showed no evidence, clinical or laboratory, of thyrotoxicosis or myxedema. In both cases a decided reduction in the metabolic rate coincided with clinical improvement.

Turnover rate of phospholipid phosphorus in the liver of the white rat, J. Bollman, E. Flock, J. Berkson (Div. of Experimental Med., Mayo Clinic, Rochester, Minn.), *Proc. Soc. Exptl. Biol. Med.* **67,** 308–313 (1948). The proportional turnover for hepatic phospholipid in the white rat is close to 5 % per hour on the average. Since the mean hepatic phospholipid concentration for the experiments was 132 mg per 100 g of liver, the mass turnover rate is about 0.8 mg of hepatic phospholipid per 100 g of body weight.

Rapid exchange of the bone salts in newborn rat as demonstrated with radioactive phosphorus, W. Percival, C. Leblond (Dept. of Experimental Surgery, McGill Univ., Montreal), *Rev. can. biol.* **7,** 217–227 (1948). Small doses of radioactive phosphate were administered to newborn rats which were sacrificed at successive time intervals from 1 to 72 hours. The distribution of radiophosphate in bones and teeth was studied by Geiger counter estimations and by autographic methods. The uptake of phosphate in all types of bone and teeth was extremely rapid suggesting that phosphate salts exchange readily and that the bony structures are being renewed at a rapid rate in the newborn rat.

Radioautographic technique with C^{14}, A. MacDonald, J. Cobb, A. Solomon (Biophysical Lab., Harvard Med.

School, Boston, Mass.), *Science* **107,** 550–552 (1948). A method is presented for taking radioautographs which provides the intimate contact required between tissue section and film and makes possible film development and staining of the section while they are in contact. The method requires that the tissue section be mounted on a glass slide. Stripping film is then cemented base side down onto the tissue, and the radioautograph is made. The film and tissue are removed together after development and cemented emulsion side down onto another glass slide permitting the tissue to be stained without affecting the emulsion.

The application of radioisotopes to problems of naval medicine, R. Smith (U. S. Naval Medical Research Inst., Bethesda, Md.) *U. S. Naval Med. Bull. Supp.* Mar.–Apr. 17–27 (1948). Experiments involving radioactive antimony with particular reference to organ distribution, exchange in animals and studies in man and in animals infected with Schistosoma are reviewed. The use of rare gas isotopes in developing a possible hypothesis for decompression sickness and deep-sea diving is presented. Also reviewed is the use of radiohydrogen in measuring body water *in vivo* in man and animal. Radiobiological trends are noted.

Preparation of gas samples for mass spectrometric analysis of isotope abundance, D. Sprinson, D. Rittenberg (Dept. of Biochemistry, Coll. of Physicians & Surgeons Columbia Univ., New York), *U. S. Naval Med. Bull. Supp.* Mar.–Apr. 82–94 (1948). Procedures for the preparation of various nitrogen compounds for isotope analysis are given. Equipment used for the preparation of these samples is illustrated. Several degradative schemes for the analysis of compounds labeled with more than one atom of the same isotope are cited.

Determination of hard radiation, including preparation of samples, W. Bale (School of Medicine and Dentistry, Univ. of Rochester, N. Y.), *U. S. Naval Med. Bull. Supp.* Mar.–Apr. 100–103 (1948). A table is presented illustrating the penetrating power of beta rays. Samples emitting beta rays are prepared so thin that no appreciable self-absorption takes place. If the sample is thick compared with the penetrating power of its beta ray the instrument will measure activity per unit weight independent of variations in sample thickness. Gamma-ray measurements and absolute calibration of counters are discussed.

Detection of intermediates, criteria of purity, M. Kamen (Dept. of Chemistry and Mallinckrodt Inst. of Radiology, Washington Univ., St. Louis, Mo.), *U. S. Naval Bull. Supp.* Mar.–Apr. 115–122 (1948). Special problems involved in the isolation of small quantities of radioactive compounds are discussed. Instances are presented where crystallization to constant specific activity did not denote purity of the compound. Adherence to crude purification procedures and failure to perform adequate control experiments may result in gross errors of analysis.

The radioautographic technique, D. Axelrod, J. Hamilton (Crocker Radiation Laboratory, Univ. of California, Berkeley), *U. S. Naval Med. Bull. Supp.* Mar.–Apr. 122–141 (1948). Problems involved in the handling of tissues such as fixing, section thickness, embedding, working with unsectioned material and dehydration are discussed. The effect of the particle used, the dose to be used, total radiation necessary, factors affecting resolution, film requisites, and film handling and developing are among the problems treated. A summary is given of four radioautographic methods.

Hazards presented by radioactive materials and how to cope with them, K. Morgan (Oak Ridge Natl. Lab., Oak Ridge, Tenn.), *U. S. Naval Med. Bull. Supp.* Mar.–Apr. 142–160 (1948). Topics covered include radiation damage to the human body, requirements for radiation survey and personnel-monitoring instruments, health physics for large scale operations, and calculation of radiation of exposure.

Dosage levels in administration of isotopes to animals and man, H. Lisco (Medical and Biology Div., Argonne National Lab., Chicago, Ill.), *U. S. Naval Med. Bull. Supp.* Mar.–Apr. 161–164 (1948). The injurious effects of radioactive isotopes will depend upon the half-life of the isotope, the type and energy of radiation or radiations emitted by the isotope, the distribution of the isotope within the body and the turn-over rate, excretion rates, and eventual retention in the body. A negative blood count is not a reliable general test

for radioactivity effects. It is usually reliable only when radiophosphorus or radioiron is used.

Laboratory handling of radioactive material; protection of personnel and equipment, P. Tompkins (Chemistry Div., Oak Ridge Natl. Lab., Oak Ridge, Tenn.), *U. S. Naval Med. Bull. Supp.* Mar.–Apr. 164–175 (1948). Techniques are presented within reach of the average laboratory which can successfully control spread of contamination and prevent overexposure to external radiation. Techniques for conducting operations at the millicurie level are tedious and inconvenient.

An illustration of the power of isotopes in a biochemical problem, V. du Vigneaud (Dept. of Biochemistry, Cornell University Med. Coll., New York), *U. S. Naval Med. Bull. Supp.* Mar.–Apr. 176–184 (1948). The problem of the mechanism of the conversion of methionine to cysteine is discussed, and several possible theories are presented. From the evidence obtained through the use of isotopes it is concluded that only the sulfur of methionine is used in the synthesis of cystine.

Medical aspects of an atomic disaster plan, E. King (Bur. of Medicine and Surgery, Navy Dept.) *U. S. Naval Med. Bull. Supp.* Mar.–Apr. 185–204 (1948). A twelve-point plan is presented which features the training of medical and lay personnel in the effects of ionizing radiation, establishment of dressing and decontaminating stations in the outlying areas of locations of potential explosions, a program of decentralization of industry, government, housing and hospital facilities, storage cites of nonperishable foods and medical supplies with adequate protection from radiation, an adequate supply of special clothing for rescue workers, and the setting up of a water supply protected from contamination with radioactive materials.

Chemical methods of isotope separation, A. Reid (Sun Oil Company, Marcus Hook, Pa.) *U. S. Naval Med. Bull. Supp.* Mar.–Apr. 205–210 (1948). A table is given listing the exchange reactions which are commercially feasible for the production of concentrates of tracer isotopes.

. . . BERNARD KANNER

PHYSICAL PUBLICATIONS

The effects of pile bombardment on uncured elastomers, W. L. Davidson (Oak Ridge National Lab., Oak Ridge, Tenn.), I. G. Geib (B. F. Goodrich Co., Akron, O.), *J. Applied Phys.* **19,** 427–433 (1948). Changes in the properties of natural rubber, butyl rubber, and polyisobutylene caused by pile bombardment were measured in terms of Goodrich plasticity and iodine number. They are explained by breaking of chemical bonds through ionization.

Intensités absolues des rayonnements émis par ^{52}V, R. Bouchez, G.-A. Renard (Collège de France, Paris), *J. phys. radium* **8,** 289–297 (1947). After neutron irradiation, samples were used to determine the number of photons emitted by measuring the ionization produced in an ionization chamber, and to determine the number of electrons as detected by a counter. Beta-gamma and beta-beta coincidence measurements were also made. A group of conversion electrons was found due to a transition from an isomeric state of energy 250 kev.

Analyse des quantités de radium et de radiothorium présentes dans une source de mésothorium sans traitement chimique, C.-P. Victor (Institut du Radium), *J. phys. radium* **8,** 298–301 (1947). The relative amounts of radium and radiothorium in a mixture were determined by using the difference in the energy of the gamma rays emitted. The gamma rays produced photoneutrons in Be or D. By using them to activate silver, the photoneutrons were detected after being slowed in paraffin.

Calcul des observations des grandes gerbes, J. Daudin (École Normale supérieure), *J. phys. radium* **8,** 301–305 (1947). The analysis of the characteristics of air showers by coincidence measurements involving a number of counters scattered over a large area is discussed from the point of view of probability theory.

Disintegration of antimony 124, I. Feister, L. F. Curtiss, *J. Research Natl. Bur. Standards* **40,** 315–319 (1948). A magnetic lens spectrometer was used to measure the emanations from 60-day half-life Sb^{124} (produced by pile bombardment of normal antimony). The gamma-ray spectrum was measured through photoelectrons from a uranium radiator. From the Fermi plot, two beta components were found (endpoints

0.57 ± 0.02 and 2.24 ± 0.05 Mev), and two gamma lines located at 0.60 ± 0.01 and 1.69 ± 0.02 Mev. The disintegration scheme is given. Remarks are made concerning the Coulomb correction for Fermi plots.

A circuit incorporating a chronoscope recorder for use with particle counters, H. D. Evans (Univ. of Edinburgh), *J. Sci. Instr.* (London) **25,** 145–147 (1948). A high-speed mechanical recorder is part of this system, which can be adapted for use following a scaling circuit. Resolving time difficulties are discussed.

Milne's cosmology and the origin of cosmic rays, E. P. George (Univ. of London), *Phil. Mag.* **290,** 240–243 (1948). Milne's theory predicts particles incident on the earth's atmosphere with velocities up to that of light, which may correspond to the primary component of cosmic radiation. An explicit expression is derived for the energy spectrum, and the number of particles is found to vary inversely with the cube of the energy.

Gamma-rays from alpha-particle reactions, D. E. Alburger (Yale Univ., New Haven, Conn.), *Phys. Rev.* **73,** 1014–1019 (1948). The energy of the gamma rays resulting from alpha-particle bombardment of Na, Mg, Al, Si, P, and S targets has been determined by finding the absorption curve in Al of the Compton recoils produced in the wall of the first of two beta counters in coincidence. Of nine gamma rays observed, six correspond to those expected from excited states of residual nuclei as determined by Q values in α-p reactions. Others may be from states of higher excitation or result from competing reactions.

Radioactive cerium and praseodymium, M. L. Pool, N. L. Krisberg (Ohio State Univ., Columbus, O.), *Phys. Rev.* **73,** 1035–1041 (1948). Disintegration schemes are given involving the following: Ce^{139}, which decays by K-capture with associated gamma rays; Ce^{141} and Ce^{143}, which emit negative electrons and a gamma ray; and Pr^{142}, which decays by β^- emission.

Fission of elements from Pt to Bi by high-energy neutrons, E. L. Kelly, C. Wiegand (Radiation Lab., Univ. of California, Berkeley), *Phys. Rev.* **73,** 1135–1139 (1948). The intense neutron beam of the 184-in. cyclotron (produced by deuteron bombardment of a thin lead target) was used to irradiate samples in an argon-filled ionization chamber, with ionizing pulses produced by recoil fission fragments used to detect fissions. A chamber containing thorium was used as a monitor. Electron collection and fast amplification served to distinguish fission-fragment ionization from background from the pulsed cyclotron beam. The yields relative to thorium increased with neutron energy and were higher for higher atomic number of the sample, Bi showing much the highest yield and lowest threshold.

Note on stimulated decay of negative mesons, S. T. Epstein, R. J. Finkelstein, J. R. Oppenheimer (Institute for Advanced Study, Princeton, N. J.), *Phys. Rev.* **73,** 1140–1141 (1948). The suggestion is discussed that negative mesons have shorter lifetimes than positive ones because of the acceleration of the decay process when mesons are bound in atomic states. Such decay is not accompanied by gamma emission, and the ratio of decays without to decays with such emission increases with Z^5, where Z is the atomic number. However, experimental evidence is against gamma emission in normal meson decay in free space.

The disintegration of antimony 124, B. D. Kern, D. J. Zaffarano, A. C. G. Mitchell (Indiana Univ., Bloomington, Ind.), *Phys. Rev.* **73,** 1142–1148 (1948). The radiations from antimony bombarded with neutrons were investigated with a magnetic lens spectrometer. The energy of gamma radiations was measured using photoelectrons from a lead radiator and Compton electrons from a copper radiator. Evidence is given for six gamma rays from Sb^{124}. The beta spectrum itself shows an internal conversion line corresponding to the lowest energy gamma ray, and a Fermi plot indicates a complex spectrum of five components with different endpoints. A level scheme is tentatively suggested.

Energies of the beta- and gamma-rays from antimony, C. S. Cook, L. M. Langer (Indiana Univ., Bloomington, Ind.), *Phys. Rev.* **73,** 1149–1152 (1948). The beta rays from the 60-day Sb^{124} activity have been investigated with a shaped-field magnetic spectrometer of high intensity and resolution. Thin backing and counter window allow low-energy measurements. Compton and photoelectrons from lead radiators determine the gamma-ray energies. The

Kurie plot of the beta rays breaks up into five components, though the basis for such analysis is uncertain. Agreement with the results of the lens spectrometer is good.

Coincidence experiments in antimony 124, E. T. Jurney, A. C. G. Mitchell (Indiana Univ., Bloomington, Ind.), *Phys. Rev.* **73,** 1153–1156 (1948). The number of beta counts and the number of beta-gamma and gamma-gamma coincidences as a function of aluminum absorber thickness were found with a thin-window and a lead-cathode counter (for beta and gamma rays respectively). An analysis resolved the beta rays into two groups (the high-intensity, low-energy groups found by other investigators). The beta-gamma coincidences indicated that there are at least three groups of electrons and that more gamma rays are associated with low-energy than with high-energy groups, in good agreement with magnetic lens spectrometer data.

X-ray yield for γ-n reactions, G. C. Baldwin, G. S. Klaiber (General Electric Co., Schenectady, N. Y.), *Phys. Rev.* **73,** 1156–1163 (1948). Yield curves normalized with an r-meter were investigated for $C^{12}(\gamma,n)C^{11}$ and $Cu^{63}(\gamma,n)Cu^{62}$ reactions as a function of maximum bombarding energy of the gamma-rays. Yields were measured with a G-M counter, and both curves show a steep rise from threshold to a maximum, followed by a gradual decrease in yield at higher energies. The maxima of the cross sections occurred at 30 and 22 Mev for C and Cu respectively. Confirmation of the decrease in (γ,n) cross section at high energies was found from absorption measurements using the photo-nuclear reaction as a detection process.

The conservation of momentum in the beta decay of Y^{90}, C. W. Sherwin (Univ. of Illinois, Urbana), *Phys. Rev.* **73,** 1173–1177 (1948). Monolayer sources of yttrium (90) give off electrons and recoil nuclei. A circuit using the electron count to start an oscillograph time base gives the number of recoils of various energies as a function of angle and recoil energy. A 6-ev correction to the observed recoil energy is made for the energy loss in escaping from the surface. Since no exact limits of error due to chemical effects at the surface can be set, the assignment of a $(1 - \beta \cos \theta)^2$ correlation function for the electron and neutrino is tentative (θ is the angle between them and β the ratio between the velocity of the electron and that of light).

Proton-proton scattering at 7 Mev, I. H. Dearnley, C. L. Oxley, J. E. Perry (Univ. of Rochester, N. Y.), *Phys. Rev.* **73,** 1290–1306 (1948). Protons of 7 Mev from a cyclotron were scattered in hydrogen gas and detected in photographic plates simultaneously for angles from 10°–45°. An experimentally determined correction was made for scattering from the defining slits. Proton energies were determined from range and magnetic field deflection. Analysis of results indicated a somewhat smaller s phase shift than did lower-energy data. The p wave analysis fits the Rarita-Schwinger symmetrical square-well tensor interaction.

Inelastic scattering of fast neutrons by Fe, Pb, and Bi, L. Szilard, S. Bernstein, B. Feld, J. Ashkin (Univ. of Chicago, Ill.), *Phys. Rev.* **73,** 1307–1310 (1948). Ra-Be and Ra-B neutrons produced fissions in a U^{238} detector. The source was then surrounded by spheres of Fe, Pb, and Bi. The ratio of the counting rates allowed a derivation of the inelastic scattering cross section, if the radius of the sphere was small compared to the mean free path for total scattering, and the elastic scattering small compared to the inelastic. The finite spread of neutron energies gave rise to a "hardening" effect as the radius of the sphere was increased. For all three elements the inelastic scattering was greater for Be than for B neutrons, and increased with atomic weight of the scatterer.

Photo-effects in middle-weight nuclei, L. I. Schiff (Stanford Univ., Stanford Cal.), *Phys. Rev.* **73,** 1311–1313 (1948). An explanation of the high ratio of cross sections for (γ,p) and (γ,n) processes in nuclei of Z from 30 to 50 is put forward, based on the statistical model. The regular level density of the residual nucleus is estimated, and ratios calculated are in good agreement with experimental results, but vary too quickly with Z.

The rate of decay of fission products, K. Way, E. P. Wigner (Oak Ridge National Lab., Oak Ridge, Tenn.), *Phys. Rev.* **73,** 1318–1330 (1948). By assuming a semi-empirical relation for the mass of a nucleus of mass number A and charge Z, as well as an approximate empirical relation between half-life and disintegration energy, a statistical approach gives estimates of the number of beta disintegrations per second and the

rate of energy liberation. These calculations assume that the light and heavy fission product chains are equally long. The average number of beta disintegrations per fission is six; the average energy of the radiations of the fission products is about 22 Mev, about a half in the form of neutrinos and a quarter each in beta and gamma rays.

The directional intensity of cosmic rays at several altitudes, M. Sands (Massachusetts Inst. of Technology, Cambridge), *Phys. Rev.* **73,** 1338–1340 (1948). A counter-telescope and coincidence circuit was used to measure the cosmic-ray intensity at various angles to the vertical and at various altitudes (in a B-29 airplane) up to 10,700 meters (pressure 250 gm/cm²). The absolute intensity at sea level of particles from the vertical which penetrate 5 gm/cm² of brass is 1.07×10^{-2}/sec/cm²/steradian.

On the analysis of extensive cosmic-ray shower data, H. W. Lewis (Institute for Advanced Study, Princeton, N. J.), *Phys. Rev.* **73,** 1341–1345 (1948). On the basis of the primary electron hypothesis and the cascade theory, expressions are derived for the number of N-fold coincidences among counters of area A at a depth of l shower units below the top of the atmosphere. Comparison with experimental counting rates gives satisfactory agreement except for variation with altitude at high altitudes (probably due to the invalidity of an approximation there).

Crystal orientation in Al by slow neutron diffraction, G. Arnold, A. H. Weber (Argonne National Lab, Chicago, Ill.), *Phys. Rev.* **73,** 1385–1389 (1948). A mechanical velocity selector was used with the neutron beam from the Argonne heavy-water pile in the region 0.003–0.05 ev to investigate crystal orientation effects by total cross section measurements.

The latitude dependence of neutron densities in the atmosphere as a function of altitude, J. A. Simpson (University of Chicago, Ill.), *Phys. Rev.* **73,** 1389–1391 (1948). Measurements were made of neutron densities and of vertical intensities of mesons penetrating 20 cm of lead and charged particles penetrating 5 gm/cm² of brass. The altitude dependence of neutron density, $D = D_0 \exp(-bp)$, where p is the pressure in cm Hg and b depends on the magnetic latitude effect, is the same as in free space. It is concluded that all or most neutrons in the atmosphere are of secondary origin.

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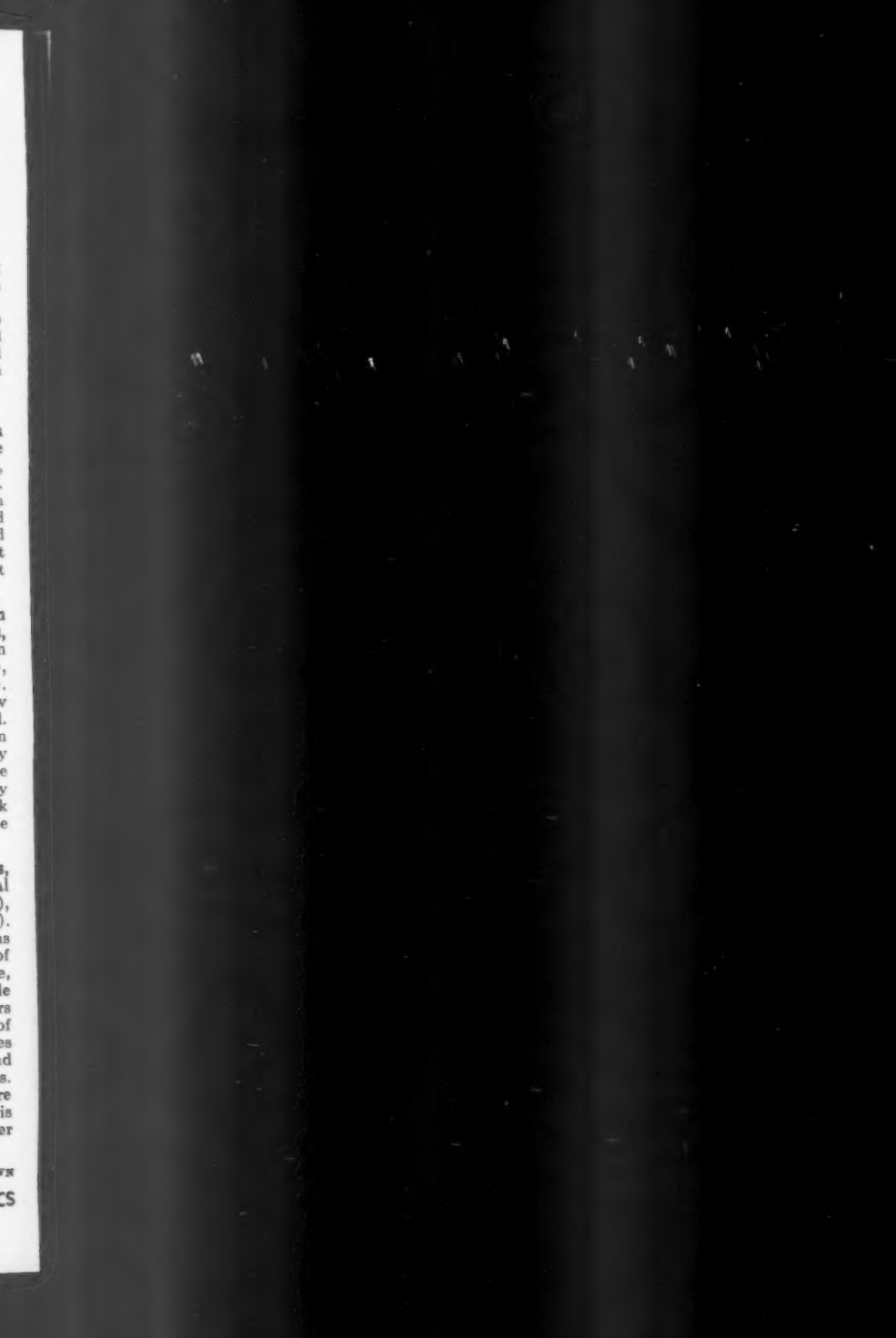
Precision wave-length and energy measurement of gamma rays from Au^{198} with a focusing quartz crystal spectrometer, J. W. M. DuMond, D. A. Lind, B. B. Watson (California Inst. of Technology, Pasadena), *Phys. Rev.* **73,** 1392–1394 (1948). A one-curie source gave a maximum counting rate five times the background (which is due chiefly to collimator scattering). The full width at half-maximum was 0.125 x.u., because of the geometrical width of the source and slight focal aberrations (natural width being much narrower). The energy was 0.4112 ± 0.0001 Mev.

Construction and performance of a low-voltage arc as a source of positive ions, S. K. Allison (Univ. of Chicago, Ill.), *Rev. Sci. Instr.* **19,** 291–298 (1948). The construction is described with diagrams. A wiring diagram included explains the operation of the arc (used in a Cockcroft-Walton generator). At 0.75 amp arc current, the beam current is 1.1 ma for a 3.5-kev probe voltage.

Alpha-particle and deuteron tracks in Eastman NTA photographic plates, R. L. Brock, E. Gardner (Radiation Lab., Univ. of California, Berkeley), *Rev. Sci. Instr.* **19,** 299–303 (1948). Alpha-particles of energy up to 380 Mev and deuterons up to 190 Mev were used. The tracks were of range greater than the emulsion thickness, so that only sections of a complete track could be studied. The variation of grain density with distance from the end of the track was plotted for particles of each type ending in the emulsion.

Self-quenching halogen-filled counters, S. H. Liebson, H. Friedman (Naval Research Lab., Washington, D. C.), *Rev. Sci. Instr.* **19,** 303–306 (1948). Small percentages (1–10%) of halogens mixed with inert gases, to a pressure of 10 cm Hg, give quenching, pulse size, and efficiency characteristics comparable with those of argon-alcohol G-M counters containing 10% of alcohol at pressures of the order of 10 cm Hg. The lifetimes of halogen counters are unlimited, and the threshold voltages as low as 250 volts. The dead time and recovery time are about the same, though build-up time is greater for halogen counters at the lower voltages.

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